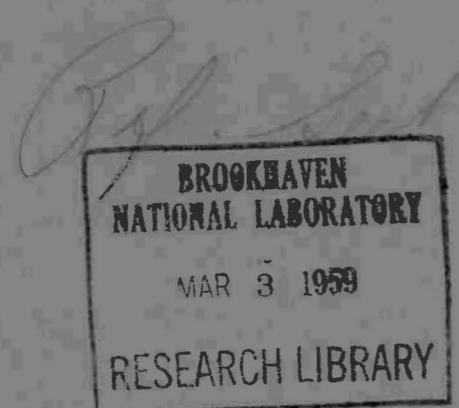


# BROOKHAVEN NATIONAL LABORATORY

*Annual Report*

**July 1, 1958**



Associated Universities, Inc.

under contract with the

United States Atomic Energy Commission

***Annual Report***

***July 1, 1958***

**BROOKHAVEN NATIONAL LABORATORY**

**Associated Universities, Inc.**

**Upton, New York**

Brookhaven National Laboratory is operated under a contract between the United States Atomic Energy Commission and Associated Universities, Inc. This, the ninth in a series of unclassified Annual Reports, gives an account of the progress of the Laboratory during the period July 1, 1957 - June 30, 1958, and its plans for the future. It is submitted under the terms of Contract No. AT-30-2-GEN-16 between Associated Universities, Inc., and the Atomic Energy Commission.

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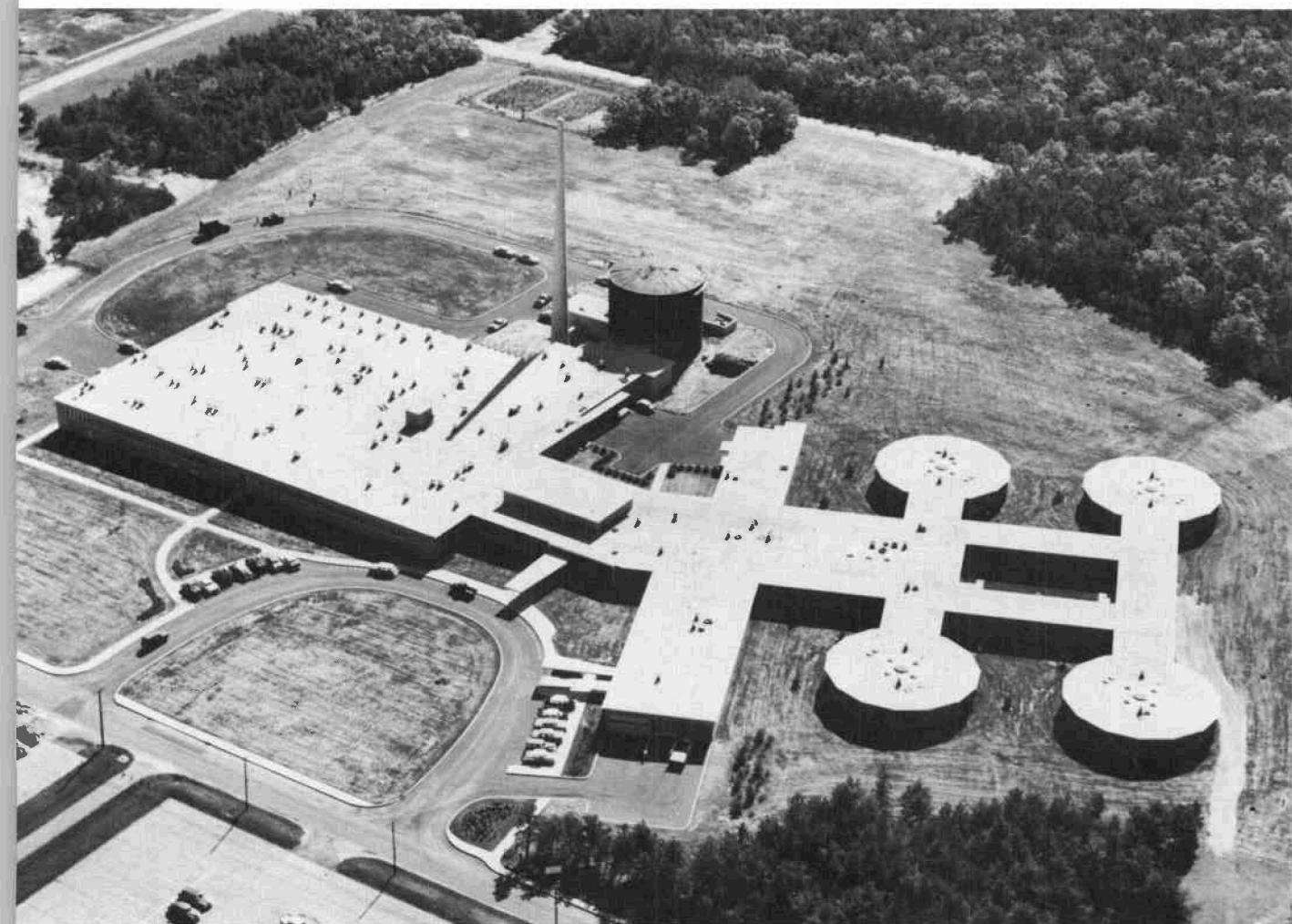
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Aerial view of the new Medical Research  
Center shortly before completion.

# Introduction

This annual report of Brookhaven National Laboratory describes its program and activities for the fiscal year 1958. The progress and trends of the research program are presented along with a description of the operational, service, and administrative activities of the Laboratory. The scientific and technical details of the many research and development activities are covered more fully in scientific and technical periodicals and in the quarterly scientific progress reports and other special reports of the Laboratory. (A list of all publications may be found in Appendix A, and the record of publications since 1946 is shown in Figure 2.)

In this introductory section may be found a general summary of the Laboratory's activities. More detailed information is given in the following sections of this report.

## LABORATORY OBJECTIVES

Brookhaven National Laboratory is a regional research center where fundamental and applied research is carried out in the nuclear sciences and related subjects as an integral part of the nationwide program of the Atomic Energy Commission by the Laboratory staff and scientists from other institutions, especially those in the Northeastern United States. It was established by a group of universities in cooperation with each other and with the Government in recognition of the need for large and expensive equipments and concentrations of scientific manpower for the successful prosecution of nuclear research. Nine leading northeastern universities (Columbia, Cornell, Harvard, Johns Hopkins, Massachusetts Institute of Technology, University of Pennsylvania, Princeton, University of Rochester, and Yale) sponsored a nonprofit corporation, Associated Universities, Inc., which, as one of its activities, administers and operates the Laboratory under a prime contract with the Atomic Energy Commission. The principal objectives of the Laboratory are

- 1) To seek new knowledge in the nuclear sciences and related fields with emphasis on programs requiring research tools not avail-

able at most universities, such as nuclear reactors, accelerators, and special laboratories.

- 2) To encourage appropriate use of its facilities by scientists from college, university, industrial, and other laboratories.
- 3) To assist the Atomic Energy Commission in the solution of specific problems.
- 4) To assist in the training of scientists and engineers and otherwise to further the dissemination of scientific and technical knowledge.

The cooperative nature of the Brookhaven program is of paramount importance. Visitors from other institutions who take advantage of the special opportunities at Brookhaven to carry out specific research and to gain useful knowledge and experience represent a significant and increasing fraction of the scientists and engineers directly engaged in the scientific program.

The objectives listed above exert a profound influence on the activities of the Laboratory and on planning with respect to both staff and facilities. The constantly changing work and the rotation of the staff demand a maximum of flexibility. For the development, construction, and effective utilization of advanced scientific equipment, the continuing presence of specially skilled groups is required, as are adequate and specialized laboratories and other facilities.

## RESEARCH FACILITIES

### Reactor

The research reactor continues to support a major fraction of the research effort at Brookhaven. This is a graphite-moderated air-cooled pile. Originally the fuel elements consisted of natural uranium in aluminum cans. During the past year all these natural uranium fuel elements have been removed, and 292 channels have been loaded with new, highly enriched ( $>90\% \text{ U}^{235}$ ) aluminum-clad fuel elements. A central flux of  $2 \times 10^{13} \text{ neutrons/cm}^2\text{-sec}$  is now available as compared with  $5 \times 10^{12}$  with the old natural uranium fuel elements, at a power level of 13 Mw as compared with  $\approx 24$  Mw previously.

The reactor is operated 24 hr a day except for shutdowns regularly scheduled every two weeks for preventive maintenance and rearrangement of experiments. An additional shutdown for a period of 24 days was required during the past year for the reloading of the reactor. There were only 12 accidental or emergency shutdowns, of which 8 were attributable to electrical power failures.

All the reactor's experimental facilities have been utilized to the greatest practicable extent for the past several years. During the past year, organizations outside the Laboratory made use of 25% of these facilities either on a rental basis, or for isotope production or other irradiations. Ten industrial organizations, 2 educational institutions, and 2 government agencies rented experimental holes for access to neutrons. The other 75% of the reactor's research facilities were utilized by various Laboratory departments.

### **Cosmotron**

The Cosmotron also normally supports an important part of the research effort at Brookhaven. Unfortunately, research with this machine was restricted to only a few months by a breakdown on November 5. Until that date the Cosmotron had been operating quite satisfactorily. Beams of 0.5 to  $1 \times 10^{11}$  protons at energies up to  $3 \times 10^9$  ev were available. The breakdown occurred in the magnet coil and was identical in nature to one that had occurred in another quadrant of the machine nine months earlier. Both breaks were attributed to fatigue hardening of the copper at places where mechanical stresses caused a slight motion. Since it was probable that similar fatigue weakness existed in numerous other members of the magnet coil, it was decided to undertake a major repair job. Examination of other magnet coil members after dismantling demonstrated the wisdom of this decision. Redesign of the magnet coils to eliminate previous defects and provide improved insulation has been completed. The delivery of these new coils has begun and should be completed in the fall of 1958.

The Atomic Energy Commission has authorized the Laboratory to proceed with a major enlargement of the experimental area and the provision of a more adequate radiation shield. The 100×180-ft structure to house the new experimental area is now being constructed as a continuation of the existing area. The new area will be serviced by a 40-ton crane and will have a floor loading capac-

ity for shielding blocks of 5000 lb/ft<sup>2</sup>. Several foci will be available for the three external proton beams to be directed into this area, and some semipermanent secondary beam arrangements are also being provided. These should greatly increase the effective use of the Cosmotron's operating time for research. Since the demand for time at this machine has always exceeded that available in a 24-hr day, the new arrangements should prove invaluable.

The new shielding blocks now under fabrication will provide sufficient thickness to keep all operating areas in the Cosmotron below the tolerance level for radiation resulting from a circulating beam of  $10^{12}$  protons per pulse, which is believed to be an attainable intensity. The long shutdown has also made possible a number of improvements in various components of the machine. It is anticipated that a completely shielded and improved Cosmotron with greatly expanded experimental area will resume operation in the first quarter of calendar 1959.

### **60-in. Cyclotron**

The 60-in. cyclotron has operated reliably throughout the past year and has been in regular use by the Physics, Chemistry, and Medical Departments at BNL and by scientists from a number of other institutions. Guests of the Laboratory from Mt. Sinai Hospital and from Columbia, Yale, Princeton, Rutgers, and Johns Hopkins Universities have used its beams on a regularly scheduled basis. More occasional use has been made by groups from MIT, the University of Pennsylvania, Livermore Laboratory (University of California), Harvard University, the Army Chemical Center, Haverford College, the General Electric Company, the Veterans' Administration, and others.

Emphasis has been placed on flexibility of operation of the cyclotron. Three different kinds of particles may be accelerated - protons to energies of  $\approx 10$  Mev; deuterons, to 20 Mev; and alpha-particles, to 40 Mev. Deflected beam intensities ranging from 50  $\mu$ a or more to  $< 0.001 \mu$ a are regularly used. Changes from one type of particle beam to another are made easily and quickly. A wide variety of target arrangements has been developed. Some special consideration has been given to the reduction of neutron background in connection with nuclear reactions in which neutrons are produced.

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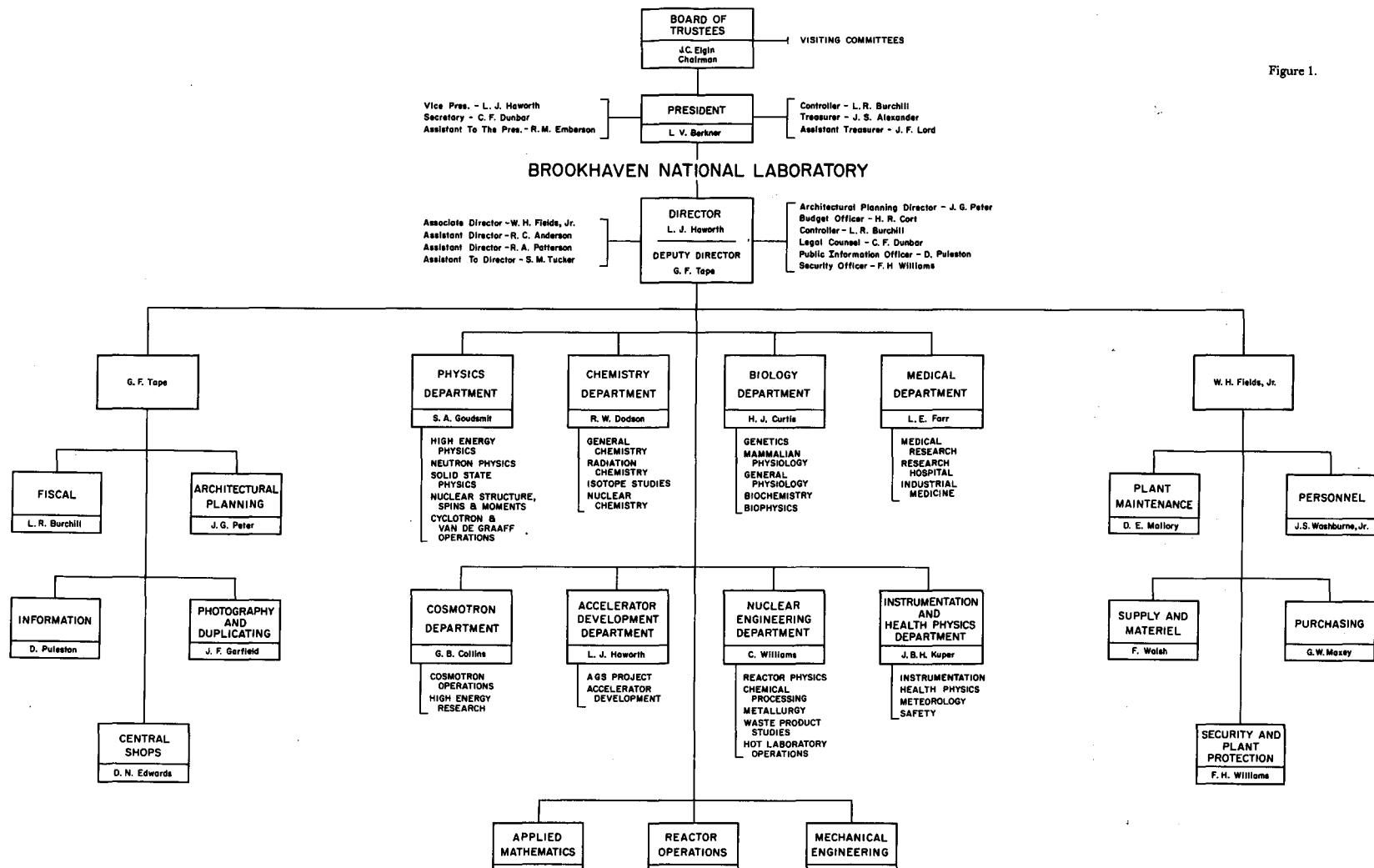


Figure 1.

Table 1

Organizational Expenditures - Fiscal 1956, 1957, 1958  
 (Includes Operating, Services to Fixed Assets, and Work for Others.  
 Direct Costs of AGS and Other Fixed Assets Are Not Included; See Table 3)

	Salaries, Wages, Insurance								Man-Years		
			Consultants & Temporary Employees	Material & Supplies	Subcontracts & Special Procurements	Power	Miscellaneous (Net)	Total Organizational Costs	% of Total	Scientific (Incl. Guests)	Others
	Staff	Travel									
Physics & Chemistry Research	1958	2,533,881	127,662	94,859	683,537	298,681	31,682	—	3,770,302	21.3	210.0
	1957	2,118,366	121,136	62,664	690,781	53,805	60,355	—	3,107,107	21.6	196.0
	1956	1,951,118	88,882	81,998	464,265	7,054	64,491	(13)	2,657,795	22.4	199.5
Biology, Medicine & Biophysics Research	1958	1,457,015	60,152	72,079	330,976	36,809	—	(8,001)	1,949,030	11.0	85.0
	1957	1,295,548	43,526	47,063	256,357	7,647	—	(5,103)	1,645,038	11.4	91.5
	1956	1,116,014	38,319	39,024	211,703	8,214	—	(15,626)	1,397,648	11.8	80.0
Nuclear Engineering Research	1958	1,425,945	32,136	51,368	426,081	1,662,173	—	—	3,597,703	20.3	83.5
	1957	1,166,905	22,579	47,588	388,640	931,284	—	—	2,556,996	17.7	81.5
	1956	907,727	19,894	43,093	374,635	190,602	—	(276)	1,535,675	13.0	77.0
Training & Education	1958	19,324	8,220	9,098	2,217	59,298	—	—	98,157	0.6	8.5
	1957	21,389	4,238	3,406	3,790	—	—	—	32,823	0.2	2.0
	1956	15,474	735	1,860	11,391	—	—	—	29,460	0.2	1.5
Radiation Protection	1958	291,109	1,284	1,858	43,421	—	—	(1,573)	336,099	1.9	11.0
	1957	265,909	1,073	1,378	46,270	—	—	(8,514)	306,116	2.1	10.0
	1956	237,004	1,768	2,141	34,268	—	—	(14,358)	260,823	2.2	14.0
Supporting Scientific & Technical Services	1958	2,066,535	16,890	18,975	346,487	69,299	420,449	(256,551)	2,682,084	15.1	29.5
	1957	1,766,212	10,277	15,405	274,860	—	496,884	(270,775)	2,292,863	15.9	27.5
	1956	1,579,781	6,705	15,066	256,976	—	470,945	(244,127)	2,085,346	17.5	29.0
Security & Plant Protection	1958	503,223	133	896	12,681	—	—	2,552	519,485	2.9	—
	1957	459,383	—	762	10,171	—	—	2,088	472,404	3.3	—
	1956	448,685	—	392	10,541	—	—	(35)	459,583	3.9	—
Miscellaneous (including Lighting, T & T, Heating Fuels, Special Maintenance, etc.)	1958	—	—	495	118,650	—	289,875	956,898	1,365,918	7.7	—
	1957	—	—	431	132,780	—	195,640	572,920	901,771	5.7	—
	1956	—	—	1,127	86,541	—	176,154	513,413	777,235	6.5	—
General and Administrative	1958	2,747,959	36,092	62,110	369,673	—	—	(109,493)	3,106,341	17.6	—
	1957	2,460,312	20,279	66,672	272,598	822	—	(150,010)	2,670,673	18.5	5.0
	1956	2,222,434	10,798	58,524	243,278	—	—	(153,256)	2,381,778	20.1	5.5
Laboratory Total	1958	11,044,991	282,569	311,738	2,333,723	2,126,260	742,006	583,832	17,425,119	98.4	427.5
	1957	9,554,024	223,108	245,369	2,076,247	993,558	752,879	140,606	13,985,791	97.0	413.5
	1956	8,478,237	167,101	243,225	1,693,598	205,870	711,590	85,722	11,585,343	97.6	406.5
AUI Administration	1958	—	—	—	—	—	158,000	158,000	0.9	—	—
	1957	—	—	—	—	—	150,000	150,000	1.0	—	—
	1956	—	—	—	—	—	150,000	150,000	1.3	—	—
Total AUI and BNL	1958	11,044,991	282,569	311,738	2,333,723	2,126,260	742,006	741,832	17,583,119	99.3	427.5
	1957	9,554,024	223,108	245,369	2,076,247	993,558	752,879	290,606	14,135,791	98.0	413.5
	1956	8,478,237	167,101	243,225	1,693,598	205,870	711,590	235,722	11,735,343	98.9	406.5
Work for Others, Direct Costs Only	1958	53,923	72	4,333	65,433	420	—	—	124,181	0.7	9.0
	1957	97,155	11,451	35,045	92,118	—	—	49,377	285,146	2.0	11.0
	1956	70,853	10,582	5,809	41,496	—	—	—	128,740	1.1	6.0
Grand Total	1958	11,098,914	282,641	316,071	2,399,156	2,126,680	742,006	741,832	17,707,300*	100.0	436.5
	1957	9,651,179	234,559	280,414	2,168,365	993,558	752,879	339,983	14,420,937**	100.0	424.5
	1956	8,549,090	177,683	249,034	1,735,094	205,870	711,590	235,722	11,864,083†	100.0	412.5

Note: Certain adjustments have been made in 1956 and 1957 costs as previously reported in order to make them comparable to those for 1958.

\*\$497,729 of this total was distributed to Fixed Assets and as services to Work for Others and Inventory.

\*\*\$447,475 of this total was distributed to Fixed Assets and as services to Work for Others and Inventory.

†\$291,410 of this total was distributed to Fixed Assets and as services to Work for Others and Inventory.

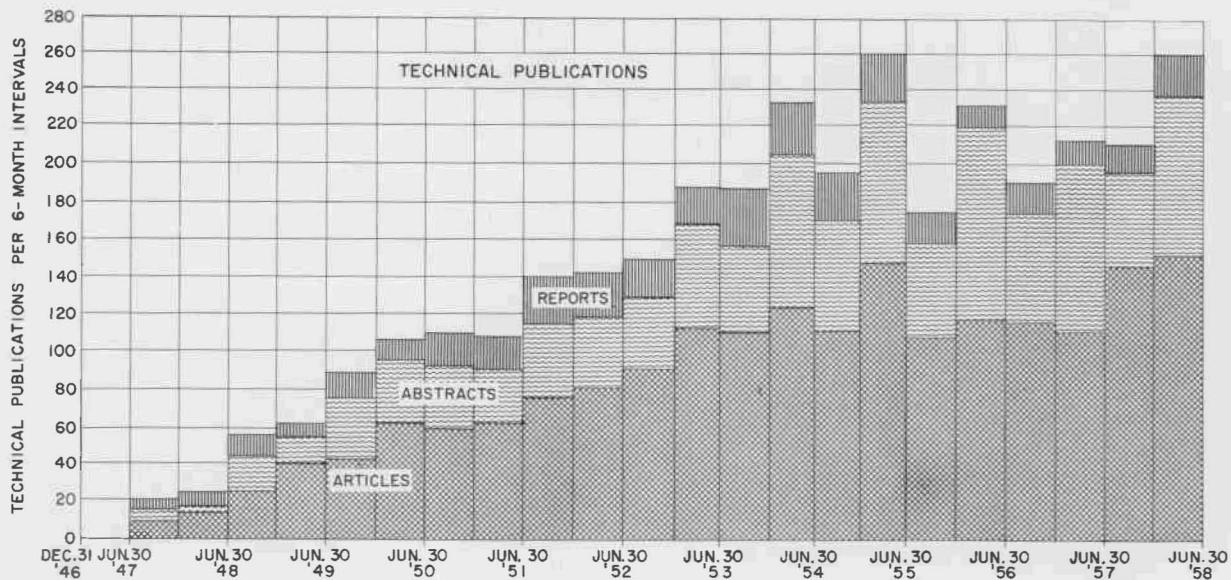


Figure 2.

Some work in which use is made of the shape of the Bragg ionization curve has been done at the 60-in. cyclotron. Since the ionization at the end of the range is considerably greater than that at the full energy, it is possible to produce tissue damage or destruction at a depth of as much as 2 mm without producing permanent visible damage or scarring at the surface. The sharpness of the ionization peak and the sensitivity of certain tissue are such that a lesion as narrow as 0.05 mm can be effected.

#### Electrostatic Generator

This accelerator of the Van de Graaff type has operated with a high degree of reliability during the past year with the continued use of the composite electrode accelerating tube developed here and described in the last annual report. A design and development program for improving the voltage characteristics of the support column structure has been initiated. It is hoped that reliable operation at voltages of  $>4$  Mev will result from this new design, as compared with 3 Mev at present. Three different particles have been accelerated: protons, deuterons, and singly ionized helium atoms. A facility for pulsing the beam of this machine has been successfully developed. All the accelerated ions are contained within bursts having time durations of 4  $\mu$ sec. The pulsed

beam may be used for experiments on the lifetimes of nuclear states or time-of-flight measurements of neutron energies.

A Van de Graaff accelerator that provides electrons at energies up to 2 Mev and the corresponding x-rays that can be produced by them is available in the Chemistry Laboratory. In general, this machine has been used in studies of the chemical effects produced by radiations.

#### 18-in. Cyclotron

The 18-in. cyclotron accelerates protons up to energies of 3 Mev, and an external beam of up to  $150 \mu$ a can be focused on a target area with a  $\frac{5}{16}$ -in. diameter at a distance of 15 ft from the machine. In April 1958 this cyclotron, which had been housed in the "test shack" in back of the Cosmotron building, was disassembled and its various components were placed in storage, since construction of the new experimental area at the Cosmotron required the removal of the test shack. An extension to the Cyclotron-Van de Graaff Building to house the 18-in. cyclotron is being constructed and should be ready for occupancy sometime next fall. Meanwhile, various modifications and improvements of the machine have been undertaken. The control system is being completely reworked, and all possible preparations are being made for the new installation.

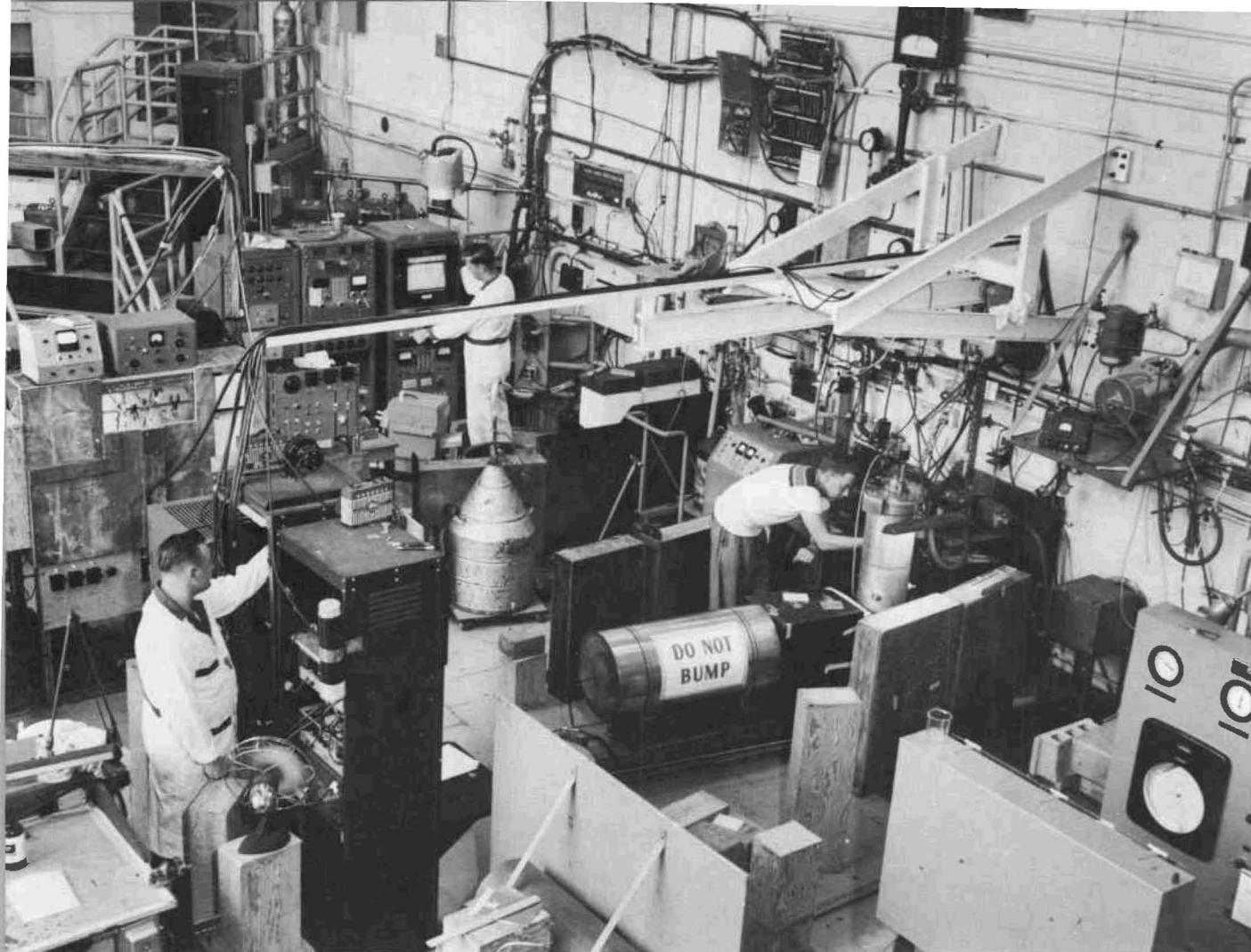


Figure 3. Experimental equipment massed at the east face of the reactor shield, showing capacity use of ports at this level.

#### CONSTRUCTION PROGRAM

The complex of structures to house the Alternating Gradient Synchrotron has been completed. This machine is a huge accelerator designed to make available a beam of protons at energies of 25 to 30 Bev. It consists of an injection system that accelerates protons up to 50 Mev and causes them to enter a circular magnetic field that keeps them moving in circular orbits 840 ft in diameter while they are being further accelerated to their final energies. The structure to house the injection system, the circular concrete tunnel  $\frac{1}{2}$  mile in circumference to enclose the magnet, and the large building surrounding the target area have been completed. All utilities, including the heating and air conditioning facilities, have been installed, and these buildings have been occupied. The steel

girders that stretch from pile cap to pile cap throughout the tunnel and which will support the magnets have been placed in position. Road construction and the landscaping of grounds around the AGS complex have been completed. The warehouse is also ready to receive deliveries of equipment and supplies.

Construction of the new Medical Research Center is nearing completion. An aerial view of this complex serves as frontispiece. The major structure contains most of the research laboratories. Their arrangement and design are based upon eight years of experience in modifying existing temporary structures to meet the needs of the Medical Department. Attached to this building is a circular tower of welded steel housing the first nuclear reactor designed specifically for medical research and therapy. The core will consist of enriched fuel elements cooled by a flow of ordinary water. Neutron moderation is provided by the cooling water and graphite blocks that surround the tank containing the core. The central neutron flux will be of the order of  $10^{13}$  neutrons/cm<sup>2</sup>-sec.

The uniqueness of this reactor lies principally in the planning of its radiation shielding. Hydraulically operated shutters, specially designed to control the neutron beams used in medical research and treatments, have been built in the concrete shielding. Forty-eight beds for research patients are provided in the four identical circular units. The location of nursing stations in the center of each unit facilitates observation of the patients and minimizes the amount of walking by the nurses in the course of their duties. Ancillary services for the hospital and space for the Clinic and the Industrial Medicine Division are provided in the building between the hospital itself and the main laboratory. The Medical Research Center is expected to be ready for occupancy sometime during the fall. Operation of the Medical Research Reactor should begin early in 1959.

The structure enclosing the extension to the experimental area of the Cosmotron, described previously under the heading "Research Facilities," is in the process of erection and should be completed by January 1959. The addition to the Cyclotron-Van de Graaff building, also described previously, which will house the 18-in. cyclotron was started in December 1957 and scheduled for

completion in August 1958. However, inclement winter weather and a labor strike have delayed work to such an extent that completion is not expected before October.

Construction of the new reclamation facility and hot laundry is well under way, and occupancy of the completed building should be possible before winter sets in. The facility will provide housing not hitherto available for the decontamination and reclamation of experimental equipment. The laundry will replace the temporary wooden building now in use for the laundering of contaminated clothing, linen, etc.

#### MASTER SITE PLAN

A long-range site and building program, known as BNL Master Plan - 1958, has superseded the 1952 Plan. In the 1958 Plan, additional Camp Upton buildings are retained for permanent use, plans for dormitories and apartments for scientists on temporary assignment are included, and some rearrangement of planned space has been made. The Plan provides a general pattern of development against which specific construction projects may be examined. Research machines are not

Figure 4. Nursing station at the center of one of the four 12-bed nursing units in the Medical Research Center. Patients can easily be observed from this central location, and the amount of walking required of nurses in the course of their duties is minimized.



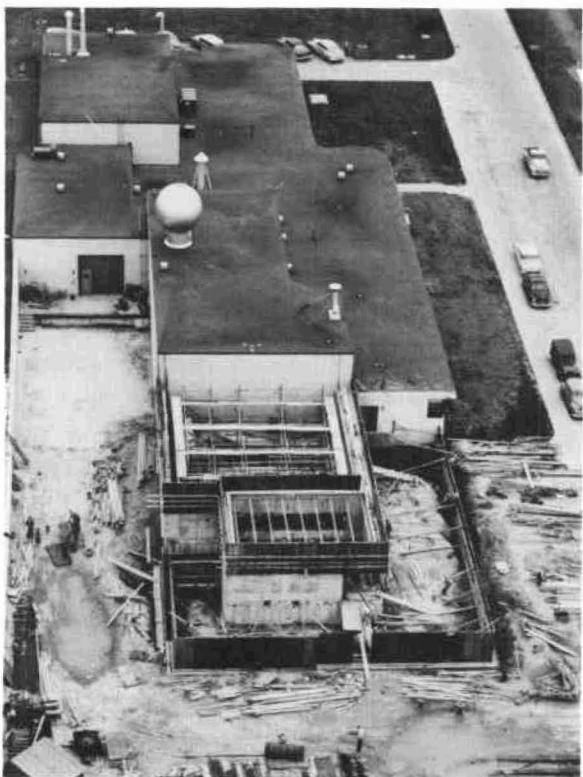


Figure 5. New 5800-ft<sup>2</sup> wing, under construction, on the Cyclotron-Van de Graaff building. When completed, the wing will house the 18-in. cyclotron.

included in this plan, since their future technological development cannot be foreseen.

Because of the nature of scientific research, long-range prediction of some of the Laboratory's requirements is not possible. This applies less to the general arrangement of the site plan and utility layouts (if they provide flexibility) than to the buildings to house the various scientific activities and services. Although the concept of the Plan as a whole will probably remain valid for several decades, some revision is likely to be required in 4 or 5 years because of changes in program emphasis and in details.

More than 150 buildings with a total floor area of  $\approx 1,300,000$  ft<sup>2</sup> are in use on the Laboratory site. These include permanent buildings (including those under construction), semipermanent buildings inherited from Camp Upton, and temporary buildings. The permanent buildings constructed since 1947 or now under construction account for a little less than  $\frac{1}{2}$  of the gross floor area. Principal among these are the buildings housing the major research machines and those for the

Biology and Medical Departments. Camp Upton buildings to be retained on a permanent basis constitute about  $\frac{1}{5}$  of the total existing floor area. These are cement block structures which will continue to serve their present purpose or will be modified under the Master Plan. About 80 temporary wooden buildings with an aggregate floor area of 440,000 ft<sup>2</sup> are in use. Because of their unsuitability for efficient operation and their increasingly high maintenance cost, the Plan provides for replacement of the majority of these. The proposed new buildings and modifications are listed below.

#### NEW BUILDINGS

Physics Building  
Nuclear Engineering Building  
Radiation Application Laboratory  
Building to house the Instrumentation and Health Physics Department and part of the Accelerator Development Department  
Critical Assembly Facility  
Plutonium Laboratory  
Chemistry Building  
Animal Quarantine Building  
Quarters for Visiting Scientists  
Administration Building  
General Services Building  
Lecture Hall and Cafeteria Building  
Central Shops Building  
Stock and Warehousing Buildings  
Plant Maintenance Buildings  
Plant Protection Building

#### EXTENSIONS

Works Area Extension for Nuclear Engineering Building  
Metallurgy Building Extension  
Biology Building Extensions  
Additions to underground utilities

#### BUILDINGS TO BE MODIFIED

Applied Mathematics Center  
AEC Area Manager and Patent Group Offices  
Part of Gymnasium Building to be converted to space for Accelerator Development Department and other engineering uses  
After the new Physics Building and Administration Building are occupied, the present Research Staff Building and Administration Building to be converted for use by the Research Library and Information Division

The cost of this program is estimated at \$38,000,000. This is a reasonable expenditure over a 5-year period for up-to-date facilities, considering that the present investment at Brookhaven is approximately \$100,000,000 (fixed asset

value after completion of construction in progress), and the annual operating budget is \$20,000,000.

## MAJOR RESEARCH PROJECTS

### Liquid Metal Fuel Reactor

The Liquid Metal Fuel Reactor Project has been under investigation at Brookhaven for several years. Both theoretical and experimental studies have been made of the feasibility of developing components that might be coordinated into a reactor system using a liquid metal as fuel. Results have indicated that in such a reactor U dissolved in molten Bi together with appropriate inhibitors can be used as a liquid fuel with graphite as a moderator. Processes have been developed to maintain a low concentration of reactor poisons in the fuel by continuous removal of such fission products. Moreover, it seems feasible to provide for breeding purposes a liquid metal blanket consisting of some form of Th in a slurry mixture using molten Bi which can also be subjected to continuous chemical processing. As a result of an engineering evaluation of the studies carried out, the Atomic Energy Commission contracted with the Babcock & Wilcox Company in November 1956 to undertake the detailed engineering design and construction of a Liquid Metal Fuel Reactor Experiment (LMFRE). The Laboratory shares the responsibility for the research and development directly required for this program and carries the prime responsibility for long-range research and development on liquid metal fueled reactors. The work of the Brookhaven staff has been closely coordinated with that of the Babcock & Wilcox Company's engineers. A brief description of the progress made during the past year by the Laboratory staff is given below. A more detailed account may be found in the Nuclear Engineering section of this report.

A neutron source reactor has been constructed to provide neutrons for the exponential studies essential for engineering design of reactor cores. This reactor was brought critical on May 19, 1958, and a few preliminary measurements have been carried out. The neutron thermal diffusion length has been measured in graphite, Bi, and Al arrays.

The process for extraction of certain fission products from the molten Bi-U fuel by means of fused salts has been brought closer to practicable application. Both the nature of the salt and metal phases involved and the mechanism of their inter-

action are better understood. Valuable quantitative thermodynamic information has been accumulated by measurements of the free energies of mixing of various solute metals in Bi and of salts in salt mixtures. An isothermal pumped salt loop made of 347 stainless steel has been operated to test mechanical components to be used for fused salts. Another larger, complex loop to provide continuous streams of salt and fuel for the final testing of salt extraction columns on a nonradioactive basis should be delivered in 2 to 3 months. Extraction with liquid Zn appears to be effective in removing those nonvolatile fission products that are more noble than Bi and cannot be slagged out by an oxidant. They form intermetallic compounds with Zn which become insoluble as the temperature is lowered and can be filtered out or skimmed off. A number of studies basic to the problem of removing the volatile products, Xe in particular, have been carried out. A bench-scale facility for studying the kinetics of Xe removal and the efficiencies of several types of degassers has been designed, and most of the construction work is completed.

The thermal convection loop program has yielded new and valuable results on the corrosion behavior of steels in contact with molten U-Bi containing Zr and Mg as inhibitors. The lower chrome steels and carbon steels are not detectably corroded in thermal convection tests. New information concerning corrosion phenomena has been obtained from studies of surface reactions between steels and liquid Bi alloys, particularly with respect to the factors that determine whether nitride or carbide films are formed on a given steel.

Nearing completion is construction of Radiation Loop No. 1, designed to study the behavior of different materials in the presence of U-Bi and suitable inhibitors. An out-of-pile run will be made this fall, and the loop will then be placed in the research reactor to determine what effects are produced by reactor irradiation.

A very large mechanical component test loop is under construction. Its main purpose is to provide a large-scale prototype for the testing of various components (including pumps, valves, pipe, pipe fittings, vessels, and instruments), as well as fabrication techniques, and system dynamics pertinent to the design of liquid metal fuel reactors. Except for radiation, this system will approximate LMFRE operating conditions. The

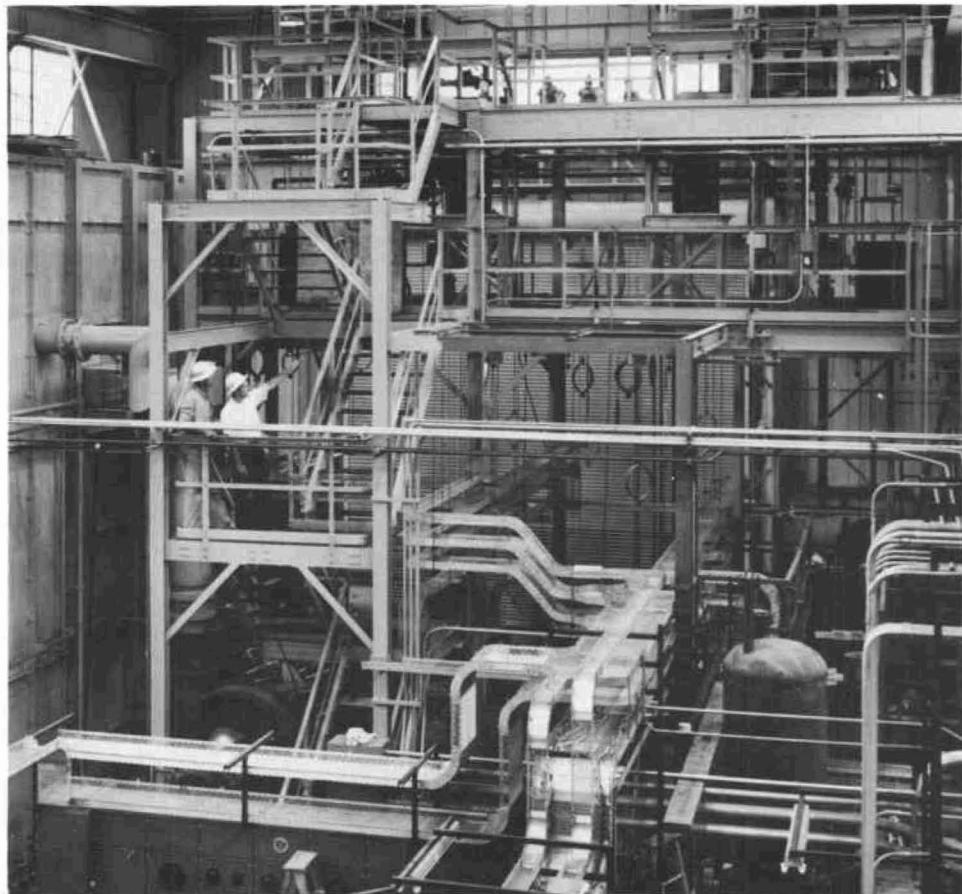


Figure 6. Interior of engineering works building, where the large-scale test loop is being assembled as part of the Liquid Metal Fuel Reactor Project. This loop will test the performance of various mechanical components and their resistance to corrosion in the presence of U-Bi solutions.

Babcock & Wilcox Company is cooperating with BNL in this project and will ultimately operate the loop.

The study of the possible use of a slurry of solid thorium bismuthide particles in liquid Bi as a fluid breeder blanket material has been continued. Viscous dispersions of thorium bismuthide can be rendered fluid by heat treatments. One subcontractor has shown that fine platelet dispersions can be obtained by gas atomization of Th-Bi solutions. Another has demonstrated that small, relatively equiaxed thorium bismuthide particles, rather than platelets, can be obtained through the use of ultrasonic energy. The mechanism by which Te additions inhibit the growth of thorium bismuthide particles is still being studied. A program has been initiated to determine the feasibility of using dispersions of insoluble Th compounds such

as the oxide, carbide, and fluoride in liquid Bi as possible fluid breeder blanket materials.

#### **Alternating Gradient Synchrotron**

The Alternating Gradient Synchrotron, already briefly described under "Construction Program," is designed to provide a beam of protons of 25 to 30-Bev energy. The development of this machine has been progressing on schedule. Contracts have been awarded for the fabrication of the main magnet coils and cores. At the end of the fiscal year 47 cores and 242 coils had been delivered; they are now being assembled. All the magnets must be subjected to rather exhaustive tests before being aligned on the steel girders now ready to receive them. Most of the switchgear, transformers, ignitrons, and auxiliary equipment for the main magnet power supply have been delivered. Fabrica-

tion of the prime mover and alternator has been delayed in order to incorporate some design changes based on experience with the Bevatron magnet power supply at Berkeley.

Initial tests of the 750-kv Cockcroft-Walton pre-accelerator were satisfactory and the machine has been moved to its final location in the linac building. Fabrication of the 10-ft-long sections of the linac tank is under way, and the assembly and contouring of the linac drift tubes are proceeding. The rf system has been designed around the immediately available French-Thomson Houston TH470 triodes. The French-Thomson Houston Company has agreed to supply the tubes, cavities, and auxiliary equipment on a satisfactory schedule.

Ferrite rings for the accelerating cavities to be located at 12 different points of the circular orbit have been received and are undergoing acceptance tests. The design of the radio-frequency accelerating system is progressing satisfactorily, and specifications for the 12 power amplifiers have been prepared to accompany the solicitations of proposals for fabrication.

Contracts for the purchase of 70 Evapor-Ion high vacuum pumps and for the fabrication of 240

sections of the magnet vacuum chamber will soon be placed.

Design of the control systems for the various linac and synchrotron components is well under way. A more detailed description of the design and construction features of the Alternating Gradient Synchrotron will be found in the Accelerator Development section of this report.

## RESEARCH ACTIVITIES

The Brookhaven research program covers a wide range of subjects in the physical and biological sciences and in engineering, but its central theme is the development and exploitation of nuclear science and technology. It can be broadly described under five major headings:

- 1) fundamental studies of atomic nuclei, the particles constituting them, and the forces involved in their structure;
- 2) study and exploitation of the physical, chemical, and biological effects of radiation;
- 3) the use of nuclear tools, such as neutrons, charged particles, gamma-rays, and isotopic tracers, in all branches of scientific research;

Figure 7. Some of the 240 magnets of the Alternating Gradient Synchrotron with coils attached prior to tests.



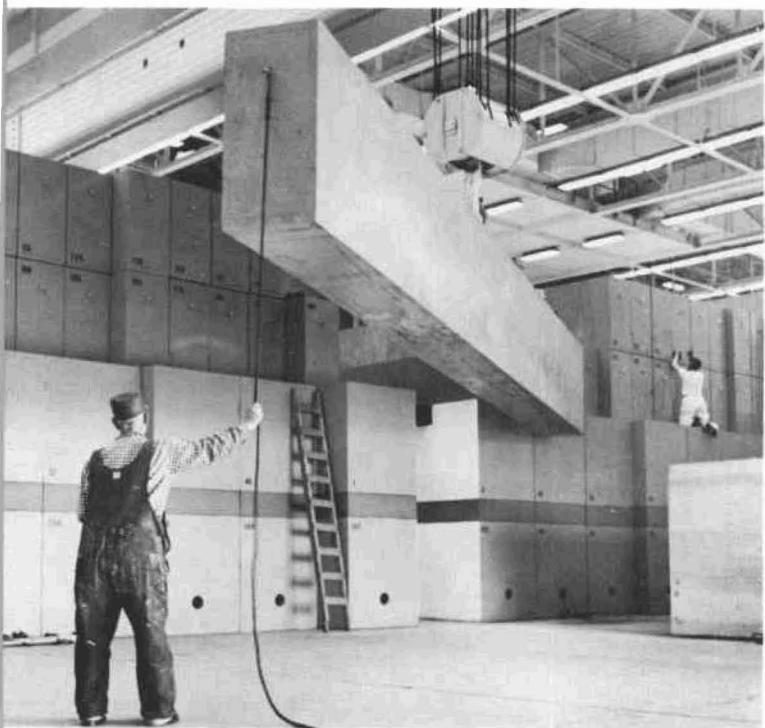
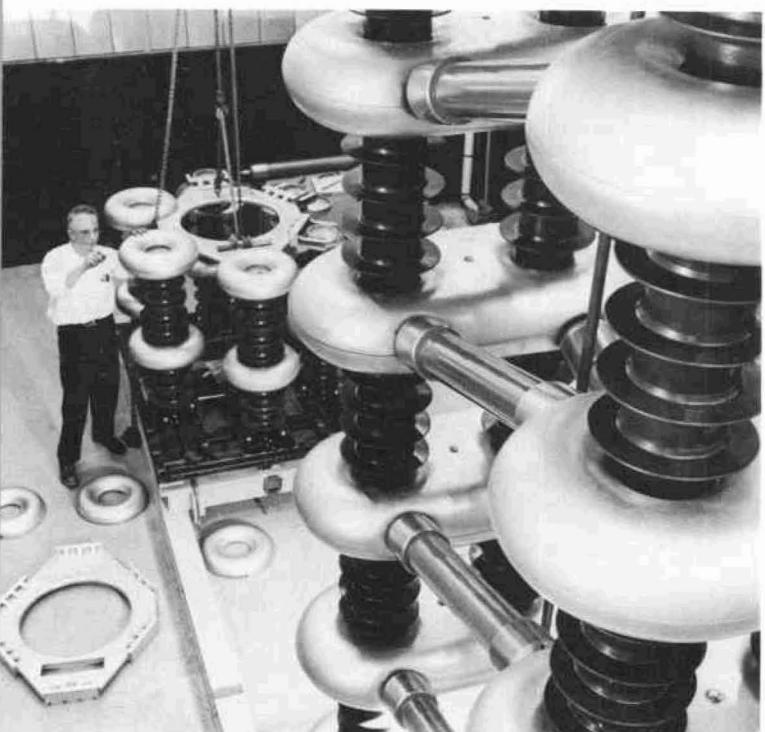


Figure 8. Assembly of high density concrete shielding blocks in the target area of the Alternating Gradient Synchrotron.



- 4) research and development not itself of a nuclear nature but useful in atomic energy development; and
- 5) useful applications of nuclear power.

A few of the more typical studies made during the past year are described below. A more detailed account of research activities is given in the next two sections of this report.

#### Fundamental Studies of Atomic Nuclei

Atomic nuclei are disturbed by means of probes, and the resulting events are studied by use of a number of different techniques. The probes range from protons of 3-Bev energy from the Cosmotron to neutrons of  $<1$ -ev energy from the reactor. The use of high energy probes usually results in the appearance of a variety of mesons and hyperons. Scientists at Brookhaven have been particularly interested in the so-called strange particles, the  $K$ -mesons and hyperons, and have established the fact that the production of a  $K$ -particle is always associated with the simultaneous appearance of a hyperon. Although the use of high energy protons was greatly curtailed this past year because of the breakdown of the Cosmotron, analysis of the data obtained has resulted in increased knowledge about the associated production of these strange particles. A number of interactions of pions with protons in liquid hydrogen bubble chambers that give rise to strange particles have been studied.

The discovery that parity is not conserved in certain weak interactions, described in last year's report, has stimulated a number of researches. For instance, it has been shown that the decay of the  $\Lambda^0$ -hyperon into a proton and negative pion violates conservation of parity. Such a violation has hitherto been demonstrated only for interactions involving the emission of a neutrino. The effect is quite large. Moreover, this reaction is not invariant under charge conjugation. However, it appears possible that the product of charge conjugation number and parity quantum number may be conserved.

The yields of  $K^+$ -mesons and of  $K^-$ -mesons when Be is bombarded by protons of different energies have been determined. Both yields fall sharply with diminishing proton energy, but the  $K^-$ -meson yield falls more rapidly. At 3 Bev the

Figure 9. The Cockcroft-Walton accelerator being assembled as part of the injection system for the Alternating Gradient Synchrotron.

yield of  $K^+$ -mesons is  $\approx 30$  times that of  $K^-$ -mesons. At proton energies of 1.7 Bev no  $K^-$ -mesons were detected, whereas the  $K^+$ -meson yield was still appreciable. These results have been shown to be roughly consistent with the principle of conservation of strangeness.

During the period preceding the Cosmotron breakdown, nuclear chemists at the Laboratory also continued their studies of the interactions of Bev protons with heavy nuclei. In particular, interactions with silver and bromine were carried out by use of emulsion techniques. Methods have been developed by which 1) reactions resulting in a recoil nucleus and an alpha-particle only are observed; 2) those in which, in addition, fragments with nuclear charge number between 2 and 6 are found; and 3) those in which two heavy fragments with nuclear charges greater than 6 have been observed. The results indicate a larger energy transfer from the proton to the nucleus for the events that result in heavy fragment emission. The abundance of low energy alpha-particles increases sharply with increasing bombarding energy. These alpha-particles have been found to be emitted in the backward direction with respect to the recoil nucleus. The frequency of events of type 3 (two fragments with atomic mass number greater than 6) increases rapidly with increasing proton energy and is about 25% of the total number of events at 3 Bev. Some of these events are fissions in which the most probable angle between the fragments appears to be about  $130^\circ$ .

An unexpected result has been found in the determination of the yields of  $\text{He}^3$  and  $\text{He}^4$  produced by the bombardment of iron with protons of energies ranging from 160 kv to 3 Bev. The yield of  $\text{He}^3$  is higher than that found earlier for tritium in this energy range. Calculations based on evaporation and cascade mechanisms have not accounted for this result.

When charged particles (such as alpha-particles, deuterons, or protons from low energy accelerators) or uncharged particles (such as neutrons or gamma-rays) interact with a nucleus, new nuclear energy states are produced which are usually unstable. Observations of the way in which unstable nuclei are produced and the manner in which they are transformed into stable nuclei yield information on the energy levels that various nuclei may occupy. The ways in which nuclei may be excited to these energy levels and the systematics of level structure in different nuclei constitute

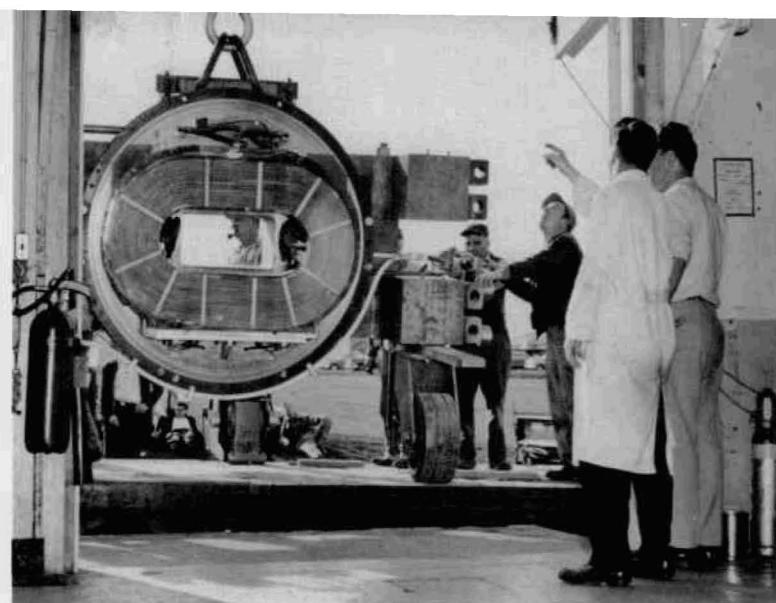


Figure 10. Rigging the magnet of the 20-in. liquid hydrogen bubble chamber.

basic data used in the formulation and testing of theories of nuclear structure. As part of a continuing program, a number of such investigations of nuclei with various numbers of protons and neutrons have been carried out during the past year. Many of them together with their theoretical significance are described in later sections of this report.

One of the consequences of the nonconservation of parity in beta decay is that the spin vector of the neutrino is either parallel or antiparallel to its direction of motion, depending upon the type of interaction which describes the beta decay. An experiment has been performed which shows that the spin and momentum vectors for the neutrino are antiparallel, that is, the sense of rotation of the neutrino is anticlockwise when viewed as it moves away from an observer. Because of its significance and ingenuity, a demonstration of this experiment will be exhibited at the Second International Conference on the Peaceful Uses of Atomic Energy to be held at Geneva in September. This experiment has also provided the starting point for both theoretical and additional experimental investigations. A number of aspects of beta decay phenomena have been clarified. Experiments to clarify others are in progress under a Columbia-Brookhaven cooperative program.

In view of the accumulated evidence that parity is not conserved in weak interactions, it became important to determine whether or not the conservation of parity is still valid in strong interactions. Three different types of experiments were carried out in efforts to detect any violation of the

conservation of parity in strong interactions. In none of these experiments could any violation of parity conservation be detected.

The well-established program of studies of neutron interaction with nuclei has been continued at the Brookhaven reactor. Many data obtained by using the fast chopper have been analyzed for the purpose of testing different theoretical models of the nucleus, especially the optical model as modified for aspherical nuclei. The agreement between theoretically predicted and measured values strongly supports this model of the nucleus. The results of measurements of "strength function" for low energy neutron scattering are in good agreement with predictions based on the aspherical modification of the optical model of the nucleus.

Crystal spectrometers have been in continuous use at the reactor to produce highly monoenergetic beams of slow neutrons and to analyze in fine detail the energy distributions of slow neutrons in cross section measurements. The high energy resolution thus attainable has made it possible to study many neutron resonances and cross sections of very narrow energy ranges. The low energy fission cross section data for  $U^{235}$  have been analyzed in terms of the one fission channel, multi-level formula developed by Reich and Moore. The results show that, even though the single channel fit is not perfect, it is sufficiently good to indicate that only a few fission channels are important.

Techniques have been developed by which short bursts of charged particles from both the 60-in. and 18-in. cyclotrons can be satisfactorily obtained. These pulses are used to produce short bursts of neutrons. Time-of-flight spectroscopic methods are then used for the study of neutron interactions at energies of a few hundred ev and in the region of 2 to 25 Mev at the 60-in. cyclotron. At the 18-in. cyclotron, measurements of the angular distributions of inelastically scattered 2.2-Mev neutrons from iron, lead, and yttrium have shown such scattering to be symmetric about 90° in the center-of-mass coordinates.

#### Effects of Radiation

Some typical studies of the effects produced by radiation on materials and on living things are described here. Results frequently reveal new information concerning the characteristics of the materials irradiated or throw new light on the internal processes in organisms.

Studies of reactor irradiation effects on various solids have been continued throughout the year. In the past very little work has been done on the dynamics of the production of crystalline defects by radiation at temperatures at which the defects tend to disappear by various annealing mechanisms, such as direct recombination of vacancies and interstitials, migration to dislocations, etc. Simultaneous production and annealing processes can now be studied by the technique of radiation-enhanced diffusion. It was pointed out in last year's report that radiation-enhanced diffusion is observable in the  $\alpha$ -brass system since it leads to increased short-range order. The phenomenon of enhanced diffusion in this system has now been studied in a quantitative way. A simple theory of radiation-enhanced diffusion has been evolved which describes the dependence of this enhancement on flux and temperature under steady-state conditions. The theoretical study also shows that the measurement of enhanced diffusion as a function of temperature can indicate the mechanism by which defects are removed from the lattice. The enhanced diffusion rate during irradiation in the Brookhaven reactor has been measured in  $\alpha$ -brass at several temperatures in the range from 0° to 190°C. This enhancement is independent of temperature in the range from 0° to 150°C, in excellent agreement with the theoretical predictions for the case when the radiation-induced defects finally disappear at internal surfaces. The data from 150° to 190°C also follow very accurately the predictions of this simple theory.

X-ray techniques and mechanical measurements have been used in investigating heavily irradiated diamond. The structure becomes highly strained, which produces an x-ray pattern characteristic of an amorphous-like structure. The hardness of irradiated diamond decreases as the total neutron bombardment increases. This effect is contrary to that in most metals and is attributed to a decrease in the cohesive energy.

Studies have been continued on the effect of reactor irradiation on single crystals of  $Al_2O_3$  by using absorption band techniques combined with annealing procedures. Simply heating  $\alpha$ - $Al_2O_3$  for several hours at 1800°C removes all the absorption in the range 2000 to 10,000 Å present in the samples when received from the manufacturer. Since this treatment doubles the transmission at 2000 Å, it is a useful and practical way of improving the ultraviolet transmission of  $Al_2O_3$  windows.

Subsequent exposure of the heat-treated samples to ionizing radiation restores the coloring to the original level.

The well-established investigation of the chemical effects in irradiated aqueous solutions has been continued, and the complexities of these systems have been greatly clarified in terms of free radical reactions. The free radicals OH and H are formed in regions of high concentration along the track of the ionizing particle; some of them combine with one another to form hydrogen ( $H_2$ ) and hydrogen peroxide ( $H_2O_2$ ), and the remainder diffuse out into the solution where they react with dissolved materials. The OH radicals are capable of oxidizing hydrogen molecules, and the product of this reaction is certainly expected to be a hydrogen atom. Experiments in which solutions containing oxygen and hydrogen peroxide as well as hydrogen are irradiated with gamma-rays show that the hydrogen atoms arising from this reaction act preferentially on the oxygen and reduce it to hydrogen peroxide, and that the hydrogen peroxide itself is not attacked by these hydrogen atoms until all the oxygen present has been consumed. However, in solutions containing only oxygen and hydrogen peroxide both  $O_2$  and  $H_2O_2$  are reduced under gamma radiation. The reducing radicals from water must act on both compounds, and the reactions have naturally been attributed to hydrogen atoms from water decomposition. It is concluded that the "H atom" arising from decomposition of water under irradiation is different from the "H atom" formed by free radical oxidation of  $H_2$ . The possibility of different forms may arise from the polarity of water which allows solutes to exist in acidic or basic forms corresponding to the loss or gain of a proton. For the hydrogen atom the acidic and basic forms would be, respectively, the ion  $H_2^+$  and the solvated electron. The relative stability of  $H_2O_2$  in acid solutions suggests that the form resulting from the oxidation of  $H_2$  may be the form more stable in acid solutions. Possibly the initial product of oxidation of  $H_2$  by a free radical is not atomic hydrogen but the ion  $H_2^+$  resulting from transfer of an electron from the  $H_2$ ; the initial radiolysis product of water could then be supposed to be a true hydrogen atom. Alternatively, the so-called H resulting from water radiolysis could be a solvated electron, while the product of oxidation of  $H_2$  would be a true hydrogen atom. Recognition of the possibility that hydrogen atoms may exist

in two forms with different relative reaction rates towards oxygen and hydrogen peroxide has clarified a considerable mass of previously published data on decomposition of water by various types of radiation.

Continuation of the study of radiolysis of pentane adsorbed on silica gel emphasizes the differences in the effect of radiation on this compound in the adsorbed state as compared to the bulk state. The formation of unsaturated compounds has been shown to be completely suppressed, but large yields of branched chain compounds were formed. The total quantity of pentane decomposed by a given dose of gamma radiation is many times greater in the adsorbed than in the bulk state. Attention has now been turned to studies of the irradiation of pentane adsorbed on compounds of the "molecular sieve" type. Results appear to show a pronounced increase in the yield of hydrogen in the adsorbed state, but no effect on the yield of methane, which indicates that certain modes of radiolytic decomposition may be strongly affected by this type of solid, while other modes are unaffected.

If an animal receives a large dose of radiation it may sicken and die within a month. If it recovers, it will still have a decreased life expectancy, due in part to the induction of cancer which does not appear until long after the irradiation, but seemingly also due to a general acceleration of the aging process. For this reason the phenomenon has come to be known as radiation-induced aging. If it can be proved that radiation does indeed accelerate natural aging, it will become a powerful tool for study of the aging process. The present program is designed not only to elucidate the basic mechanism of radiation-induced aging, but to relate it to the phenomenon of natural aging.

A current theory postulates that aging is an accumulation of the effects of stresses, each causing a certain amount of irreparable damage. Nearly two years ago a series of treatments was started in which noxious agents were administered to mice either weekly or bi-weekly for almost the entire life span. The agents used were typhoid toxin, typhoid toxoid, tetanus toxoid, tetanus toxin, nitrogen mustard, and turpentine. Provided the treatments were not so drastic as to kill acutely, the mice lived as long as the untreated animals. Hence it is necessary to look for some other mechanism by which radiation shortens the life span.

It has been previously reported that whole-body irradiation of rats affects their carbohydrate metabolism. The increase in blood glucose and liver glycogen is mediated in large part through the pituitary and adrenal glands. Removal of the pituitary or the adrenals prevents these effects.

More recent experiments show that intact rats invariably have an increased nitrogen elimination after irradiation. Adrenalectomized animals show no such increase. It appears probable that protein from the radiosensitive tissues, e.g., the spleen and thymus, is transferred in part to the liver as amino acids that are converted to glycogen, the resulting nitrogen being excreted in the urine. In the absence of the adrenal glands the amino acids appear to be stored as such or as protein.

The gross damage by irradiation to the intestine in animals is well known and constitutes a prime cause of death within a certain range of dose. A study of the effects of radiation on intestinal epithelium has shown that the initial effect is to decrease cell production and reduce the cells lining the intestine to a thin layer that may persist for some time. This layer, however, is not sufficient to prevent passage of bacteria and later death. Further information has resulted from collaborative work at Brookhaven with members of other institutions, in which thymidine was labeled with tritium, the location being detected by autoradiography. The irradiation prevents resting cells from entering the phase during which desoxyribose nucleic acid (DNA) is doubled. Cells in the phase of synthesizing DNA are inhibited, and some that pass through it are unable to divide. Inhibition by radiation of the process leading to cell division (mitosis) is much more profound than the inhibition of nucleic acid synthesis.

The cooperative radiation plant mutation program instituted several years ago has been continued. Plants, seeds, cuttings, and the like are subjected to gamma, x-ray, or neutron irradiation for eventual study elsewhere, in an effort to assess the value of such irradiations in producing useful mutations. The cooperating group at Rutgers University has found a late-ripening branch in the important peach variety Elberta and a mutation from the freestone to clingstone condition in the variety Brackett. Embryos produced by certain controlled crosses of peaches are often of low viability. Of those which can be made to survive, many produce stunted plants which grow in an abnormal manner. Radiation was used in an at-

tempt to restore normal growth. The use of x-rays proved ineffective, but the Rutgers group has reported that embryos irradiated with neutrons have produced normal, rapidly growing seedlings. The effect appears to be due to a release of inhibitory factors by the neutron irradiation.

Last year the discovery that there are very marked delayed effects in seeds irradiated with x-rays was reported. Indeed, under certain circumstances as much as 95% of the damage initiated by the x-rays may develop in the period following the irradiation. This delayed effect has been found to be greatly dependent on the moisture content of the seeds at the time of irradiation. Evidence has been accumulated indicating that two processes are involved. Both are stopped immediately by the addition of water. One acts rapidly, and oxygen is required in the water used for germination in order to obtain the ultimate damage. The other acts slowly, and its effect is independent of oxygen. The total delayed effect appears to be due to an indirect reaction, that is, the radiation acts on the water molecules present in the seed to produce free radicals, which in turn react over a considerable period of time with sensitive sites within the cell to produce the damage. An effort is under way to detect the free radicals by a nuclear magnetic resonance technique to determine whether they do indeed continue to exist as long as the delayed effect continues. There is no such effect when seeds are irradiated with neutrons.

The Medical Department has continued its studies of the effects of neutron irradiation of patients afflicted with the brain tumor glioblastoma multiforme. In this neutron capture therapy a boron compound is injected into the patient prior to irradiation. Neutrons captured by boron atoms in tumor cells produce a nuclear reaction that results in an alpha-particle and a recoiling lithium atom whose energies are sufficient to be lethal to the tumor cells. During the year, in giving experimental treatments at the graphite reactor to a third series of patients, a sharply modified procedure was used. The troublesome skin lesions hitherto experienced were successfully avoided. The response of the patients, who were deliberately selected from among those in the most advanced stages of the disease, suggests that the retardation of tumor growth was greater than any previously attained. In experiments with a transplantable, highly invasive brain

tumor in mice, the tumor was rendered nonviable in more than 50% of the animals by the neutron capture technique.

Studies have also been continued of the cancerogenic effects of whole-body irradiation with x-rays in female rats. Incidence of breast tumors, measured 11 months after exposure, rises linearly with dose between 23 and 400 r of 250-kv x-rays. Ovariectomy preceding or following exposure reduces but does not eliminate the incidence of breast tumors. Intact males respond to the same degree as ovariectomized females, and castrate males give a response intermediate between that of intact males and intact females. Thus, the maximum induction of breast tumors in these animals was affected by both the level and type of gonadal hormones present. Four distinct types of breast tumors have been observed, as well as some mixed types. Shielding experiments have shown that neither the pituitary nor the ovary need be irradiated for induction of breast neoplasm.

Animal studies on the therapy of the hemopoietic syndrome of acute whole-body radiation injury have produced interesting results. Twenty dogs were given 400 r of x-radiation; of these half received therapy and half did not. Eight of the treated animals survived as compared with one in the untreated group. The therapy consisted of antibiotics (to control infection), fresh whole blood transfusions, parenteral fluid infusions, forced oral feeding, and meticulous nursing care. In the surviving treated dogs the platelets, leucocytes, and red blood cells did not return to their preirradiation values until 60 to 90 days following irradiation.

In another study of animals rendered thrombocytopenic by whole-body irradiation, a semi-quantitative measure of the degree of bleeding was obtained by enumeration of red blood cells in thoracic duct lymph. After exposure large numbers of cells were observed in the lymph. Administration of lyophilized platelets neither increased the recipients' circulating platelets nor decreased the number of red blood cells in the lymph, while administration of fresh platelet preparations did both to significant degrees.

#### Use of Nuclear Tools

Extensive use continues to be made of all available nuclear tools of research. Only a few of the numerous studies can be described here. Both physicists and chemists have been interested in

the arrangements of atoms, ions, and magnetic spin systems as revealed by the diffraction of low energy neutrons from the reactor. Studies of chemical reactions and of biological processes in plants and animals, including humans, have been pursued through the use of radioactive isotopes as tracers.

Much of the neutron diffraction program has been concerned with studies of the relationship between the arrangement of atoms or magnetic centers in crystals and their magnetic, ferroelectric, or other physical properties. Final refinement of the ferroelectric Rochelle salt structure has confirmed the structure suggested last year. Studies of other ferroelectric crystals are being made, in some cases with the cooperation of a group from Pennsylvania State University. The study of short-range order in the brasses has been complicated by the similarity of the scattering powers of copper and zinc for both x-rays and neutrons. Measurements of the scattering by  $Cu^{63}$  and  $Cu^{65}$  give values different from those obtained from natural copper. The  $Cu^{65}$  isotope has the larger scattering length, and the short-range order scattering in brasses with  $Cu^{65}$  as the copper constituent is  $6\frac{1}{2}$  times more intense than that with natural copper. Because of this, the order-disorder transformation in  $\beta$ -brass has become experimentally accessible to neutron diffraction techniques. Analysis of the data thus obtained has not been completed.

The cold neutron facility described in last year's report is being used for scattering measurements in a series of metallic hydrides which might prove useful as moderator materials in reactors. In the thermalization of neutrons by hydrogenous materials the hydrogen can be treated essentially as free for neutron energies  $>0.5$  ev. For energies lower than this, the crystal binding of the hydrogen markedly influences the energy exchange. In a cooperative program with the General Atomic Division of the General Dynamics Corporation, an investigation in this low energy region has been made of water, ice, Lucite, polyethylene, zirconium hydride, and magnesium hydride. The metal hydrides show remarkable fluctuations in the energies given up by neutrons, which can be related to the energy levels of the hydrogen oscillators in these substances.

Rare gases are produced in iron meteorites by the action of cosmic rays. The relative rare gas contents found in these meteorites are related to the relative probabilities of the production of the

same gases in iron. A joint study of the rare gas contents of iron meteorites and their probabilities of production in iron bombarded with protons from the Cosmotron should provide information on such matters as the constancy of cosmic radiation in space, the sizes of meteorites before entering the earth's atmosphere, and the length of time the meteorites have been in space. Answers to such questions should be suggestive of the origin of meteorites and possibly of the solar system itself. An unexpected result of measurements obtained

a factor of 2 or 3. It is concluded that these meteorites have not all had their present size for the same length of time, which indicates that asteroids or other sources of meteorites are continually breaking into smaller pieces.

Ages of stone meteorites have been determined from measurements of K/A ratios, lead isotope ratios, Rb/Sr ratios, or U/He ratios. These different methods lead to a maximum age of  $5 \times 10^9$  years for stone meteorites. Although the concentrations of elements which may be used for age

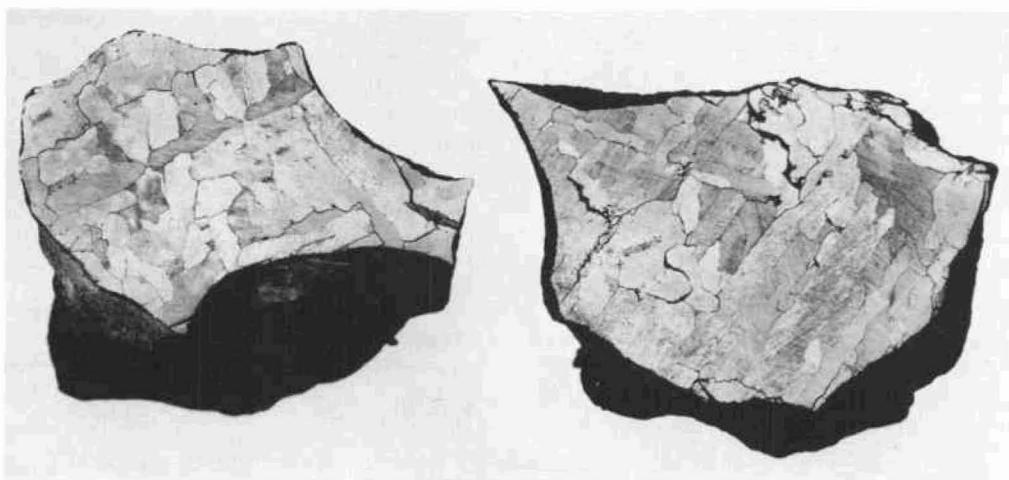


Figure 11. Two fragments of the Cañon Diablo meteorite from the large crater near Flagstaff, Arizona, which were used in determining cosmic-ray exposure ages.

of the production of helium and tritium in iron is the indication that the probability of  $\text{He}^3$  production is much higher than that of tritium in the proton range from 0.16 to 3.0 Bev. Current theoretical considerations do not account for this experimental result. Similar measurements of the production of  $\text{A}^{36}$  in iron by high energy protons have shown that 80% of the  $\text{A}^{36}$  in meteorites is the result of beta decay of  $\text{Cl}^{36}$  produced by cosmic rays; hence the  $\text{A}^{36}/\text{Cl}^{36}$  ratio should be a reliable index of the cosmic-ray exposure ages of meteorites.

Measurements have been made of the neon, argon, and helium contents in seven iron meteorites, and estimates have been obtained of the average cosmic-ray energy to which each has been subjected. These energies range from 0.5 to  $>3.0$  Bev. The cosmic-ray exposure ages deduced range from a few million to over half a billion years; although crude, they should be correct to at least

determination are much smaller in iron meteorites than in stone meteorites, neutron activation analysis is such a sensitive tool that it is possible to determine the potassium and argon contents of such meteorites. Large variations in the concentrations of potassium and argon were found in different samples of the same meteorite, but the ratio  $\text{A}^{40}/\text{K}$  was somewhat more constant. The ages obtained for these iron meteorites vary from 5.3 to  $13 \times 10^9$  years. The higher values are considered to be the more significant. The time elapsed since solidification is thought to be roughly twice as great for iron as for stone meteorites. This is consistent with the concept that the parent body of the meteorites was composed of a silicate phase and an iron phase. Since most of the radioactivity would be concentrated in the silicate phase, it would have a thermal history different from that of the iron phase.

During the year the behavior of cells under a variety of circumstances has been studied by a technique involving tritiated thymidine labeling and autoradiography. In preparation for division a cell synthesizes duplicates of certain compounds. At division each new cell contains one of the duplicates. Of particular interest is deoxyribose nucleic acid (DNA), a major constituent of chromosomes. The DNA of the chromosomes appears to be primarily involved in heredity, the process by which specific patterns of growth, metabolism, and function are transmitted to progeny. By growing cells in the presence of tritiated thymidine, a precursor of DNA, the DNA synthesized is tagged with tritium and can then be located by autoradiography. Microscopic examination of the cell and the apposed developed photographic emulsion serves to determine the part of the cell bearing the activity. Moreover, this label may be followed as cells divide or proliferate.

This same technique has been used to follow the pattern of organization of the chromosomes in the salivary glands of growing *Drosophila* larvae. Larvae from hatching eggs were fed on a medium containing labeled thymidine. At intervals larvae were removed and allowed to continue growth on an unlabeled medium. When the larvae were mature the salivary glands were removed, appropriately prepared, and covered with photographic film. Microscopic examination of both developed film and apposed glandular material followed. The findings indicate that as these chromosomes develop, new DNA appears on a strand parallel to the long axis of the chromosome, that these strands remain intact during succeeding replications through a number of endomitotic cycles, and that the DNA is not distributed uniformly along the chromosomes but is concentrated in certain regions. This constitutes relatively direct evidence for the multistrand concept of these chromosomes in mature larvae.

Tritiated thymidine and radiographic techniques have also had immediate application in the study of the genesis of the various cells of the blood. Patients with malignant diseases, but no abnormality in the blood-forming function, have been administered small doses of tritiated thymidine. The results obtained are similar to those obtained on dogs and rodents. Within 15 minutes after intravenous injection, labeling is practically completed. Labeling of large cells of coronal and central areas of the lymph follicles, cortex of the

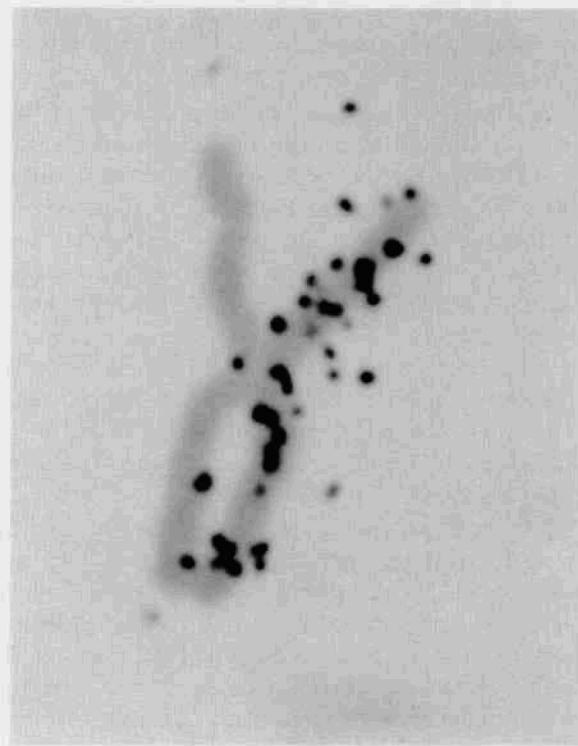


Figure 12. A chromosome labeled with tritiated thymidine. The black dots are silver grains in the emulsion which show the site of incorporation of the labeled thymidine. Note that half of the chromosome is not labeled.

thymus, white pulp and follicles of the spleen, and undifferentiated cells of bone marrow is striking. This group of cells is regarded as the primitive proliferating pool for the purposes of this study, and the evidence strongly indicates that it constitutes a common progenitor pool for all lines of blood cell formation. The intense proliferation of these pool cells, widely distributed throughout the body, was unexpected. Their turnover rate may exceed that in any other tissue.

Incorporation of label in neoplastic cells in the marrow of patients with multiple myeloma or chronic lymphatic leukemia was found to be rare, nor would the leukemic cells in the blood of chronic lymphatic leukemia incorporate thymidine. Labeling was extensive, however, in the marrow and blood of patients with acute and chronic myelogenous leukemia. The marrow of patients with infectious mononucleosis showed intensive labeling of primitive stem cells, and an increased number of labeled large mononuclear cells were present in the peripheral blood. The value

of these differences in establishing diagnosis, therapy, and/or prognosis requires further investigation.

A similar technique has been used in an attempt to gain information about ribose nucleic acid (RNA) and the nucleolus in the root tip cells of the English broad bean. The nucleolus is associated with the nucleus of cells, but, unlike the chromosomes, it contains RNA, which appears to be implicated in protein synthesis, differentiation, and embryonic development. Tritium-labeled cytidine was used to label RNA. Cytidine is known to be a precursor of both RNA and DNA. The label appeared first in the nucleolus and later in the cytoplasm. When present in the nucleus it was associated with DNA, elsewhere with RNA. When cells with only the nucleoli labeled were grown in the presence of unlabeled cytidine, the amount of label in the nucleoli decreased while that in the cytoplasm increased. The most likely explanation appears to be that RNA is formed within the nucleolus and after synthesis, or accumulation, is transferred to the cytoplasm.

Muscular contraction is known to involve the contractile protein actomyosin and transfer of energy from adenosine triphosphate (ATP), but the manner in which the energy is transferred has eluded detection. By use of water labeled with the oxygen isotope O<sup>18</sup>, a major clue to the mechanism of this conversion of chemical energy to mechanical work has been provided. When a contracting preparation of actomyosin was treated with ATP in oxygen-labeled water, observation of the pathway of O<sup>18</sup> indicated that the exchange of energy took place between the water and an intermediate formed during hydrolysis of the ATP. Evidence was obtained that the intermediate was a phosphoprotein on the myosin portion of the actomyosin, and that actin reacted to displace the inorganic phosphate from the phosphomyosin intermediate. This chain of events is consistent with the picture of contraction of striated muscle obtained with the electron microscope.

A large number of metabolic pathways were studied during the past year. Many of these are described in the Medical Research section. Considerable effort has been expended on the study of inorganic salts because of the presumed simplicity of their relationship to the systems involved, but in the studies being made this simplicity is not apparent. In a study of the uptake and retention of minerals by bones in rats, the accretion rate per

unit ash was highest in the ribs and lowest in the incisors. In an effort to determine the effect of radiation on these rates, it was found that after 800 r of partial body irradiation the accretion rate decreased, but the rate of turnover of bone-fixed strontium was unaffected by 600 r. Further studies are in progress.

Work has continued on the charting of the metabolic pathways of manganese. The high specificity of these pathways has been previously reported. Elution from the body of radioactive manganese cannot be accomplished with magnesium, transition group metals, or members of the seventh group of elements. In contrast to other elements, excreted manganese is entirely bile secreted. It is not known whether this high specificity is due to the compound with which it may complex or to the specific loci within cells where it may largely be found. Data suggest that mitochondria may constitute a major location. It seems likely that unidentified organic compounds play a significant role in delimiting specific pathways for manganese.

The observation that parkinsonism develops following manganese intoxication led to the initiation of exhaustive studies on patients with Parkinson's disease to determine whether or not their bodies handled manganese in some unusual manner associated with the disease. This study has been expanded to include patients with Friedreich's ataxia and Wilson's disease. Studies on patients have been supplemented by experimental observations on rats. The results increasingly suggest that differences in the handling of manganese may indeed exist between specifically diseased individuals and those free of this type of disturbance of the central nervous system, but further investigation is required to confirm this.

#### **Related Research and Development**

Research and development not necessarily nuclear in nature but pertinent to the development of the atomic energy program have been carried out primarily by engineers. Many of these studies have been required for the design and development of reactor components and systems, particularly in connection with the Liquid Metal Fuel Reactor. These have already been briefly described under the heading "Major Research Projects." Additional activities include studies of graphite and of fuel element development and reprocessing, the development of special chemical analyses, and the design of special instruments

for measuring radiation and of automation devices.

One of the major problems in reprocessing fuel elements by volatilization is the removal of the large quantities of heat. Because of the low heat capacities of the reactant gases, liquid carriers have been used. Recent work here has indicated that a gas-fluidized bed of granular solids may have desirable advantages.

The feasibility of using a fluidized bed as a mobile fuel for a nuclear reactor is being investigated. Such a bed might consist of uranium oxide particles suspended in liquid Na inside tubular elements. A simulated system consisting of glass beads in water has indicated that from a mechanical standpoint the suspension can be stable, uniform, and "fluid."

Following the Windscale incident in England, in which certain radioactive substances were released during a fire in the graphite moderator of a reactor, an investigation of the burning characteristics of graphite was initiated. Tests have shown that self-sustained burning can be attained if the graphite reaches 900°C. It is a function of the air velocity at this temperature, but independent of whether or not the graphite is irradiated. Above 1000°C burning is independent of air velocity, and test fires had to be extinguished by suffocation. Release of stored energy over a wide range of temperatures was also measured. Samples of graphite taken from the Brookhaven and Oak Ridge reactors were first annealed at 100°C, and then the release of stored energy was measured as temperatures were increased from 300° to 800°C. Although energy continues to be released over the range investigated, the magnitude is not great enough to cause appreciable temperature excursions.

Development of special methods of chemical analysis has been required for many research projects. Examples include the determination of trace amounts of Th in Bi and of small amounts of Al in steel containing Ti, the coulometric determination of U and Mo in the presence of each other, polarographic determination of U in Bi without prior separation, analysis of ternary salt eutectics, the determination and extraction of a few ppm U in the same eutectics, and colorimetric determinations of Mg and iodides.

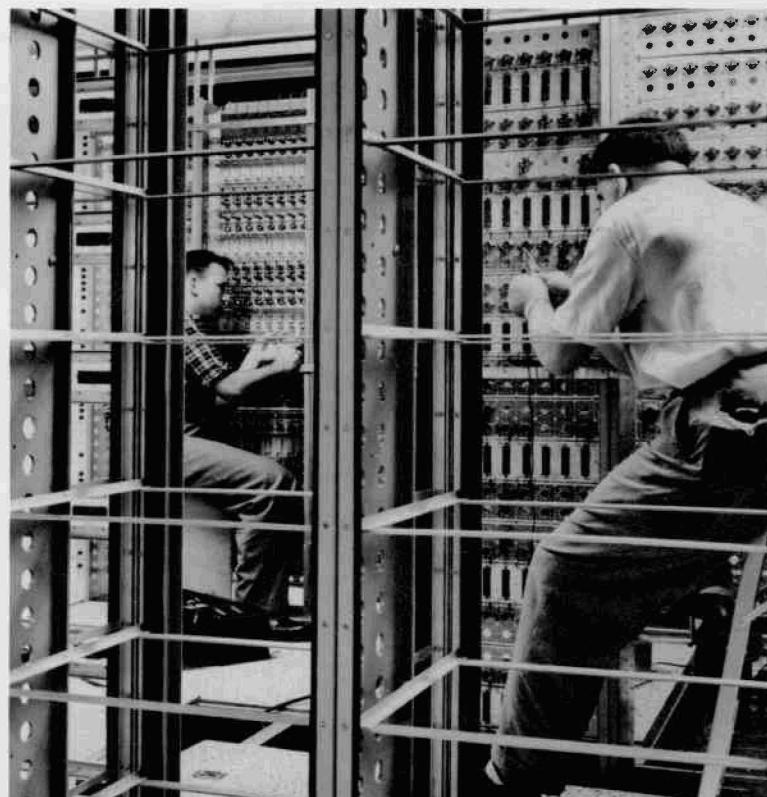
The need for computing and automation devices in carrying out the various research programs continues to increase. Substantial progress has been

made on the high capacity digital computer started in 1956 and now called Merlin. Its design is based on that of the Maniac II at Los Alamos, but modifications have been introduced that increase the high speed memory capacity and that will better serve the needs of the Brookhaven program. Members of the Applied Mathematics Division have been actively cooperating in these modifications and have worked out the order code and logical design of the control section. About  $\frac{1}{3}$  of the electronic circuits have been built.

Three major projects to provide automation devices for research scientists are also in progress in the Instrumentation Division. Such devices may be used to record data automatically and at a much faster rate than is possible manually, and even to sort and correlate the data for further analysis. One such device is designed to analyze two simultaneous radiations detected in separate detectors. The instrument will record the spectrum of pulses occurring in one detector in coincidence with each value of pulse height occurring in the second detector. It will use about 1500 transistors, have a variety of displays on a cathode-ray tube, and include provision for printing the data.

The Compilation Group at Brookhaven has prepared a second edition of BNL 325, *Neutron Cross Sections*. The first edition of this report, prepared for the International Conference on the Peaceful Uses of Atomic Energy at Geneva in

Figure 13. Assembling the high speed digital computer, the Merlin.



1955, has become the world-wide standard reference handbook of neutron cross sections. Copies of the second edition will be distributed at the Second International Conference to be held at Geneva in September. The data were obtained from laboratories in many countries throughout the world as well as from all the laboratories in the United States. It is expected that, like the first edition, this new edition will become the primary sourcebook for data on neutron reactions throughout the world.

#### Useful Applications of Nuclear Power

The Laboratory's activities in the development of nuclear reactors for power purposes have been centered for several years on investigation of the feasibility of using nuclear fuel in a liquid metal solution. The use of such a fuel for power purposes should have several advantages. A liquid metal fuel could be circulated to provide direct heat transfer and would be susceptible to continuous degassing and chemical processing. Radiation

damage to fuel would be nonexistent. High fuel temperatures would give high thermal efficiencies without excessive pressures and the attendant containment problems. A breeder blanket of similar liquid metal could also be subjected to continuous processing and might provide as much fissionable material as is consumed. Continuous processing and replenishing of fuel could be expected to keep fuel costs and inventory low and would eliminate any necessity for the shutdowns now required for fuel replacement. The activities involved in investigating these various aspects have constituted the Liquid Metal Fuel Reactor Project, already described briefly under the heading "Major Research Projects." Further technical details will be found in the Nuclear Engineering section of this report.

#### PERSONNEL

The total number of people on the Laboratory payroll on June 30, 1958, was 1828 exclusive of

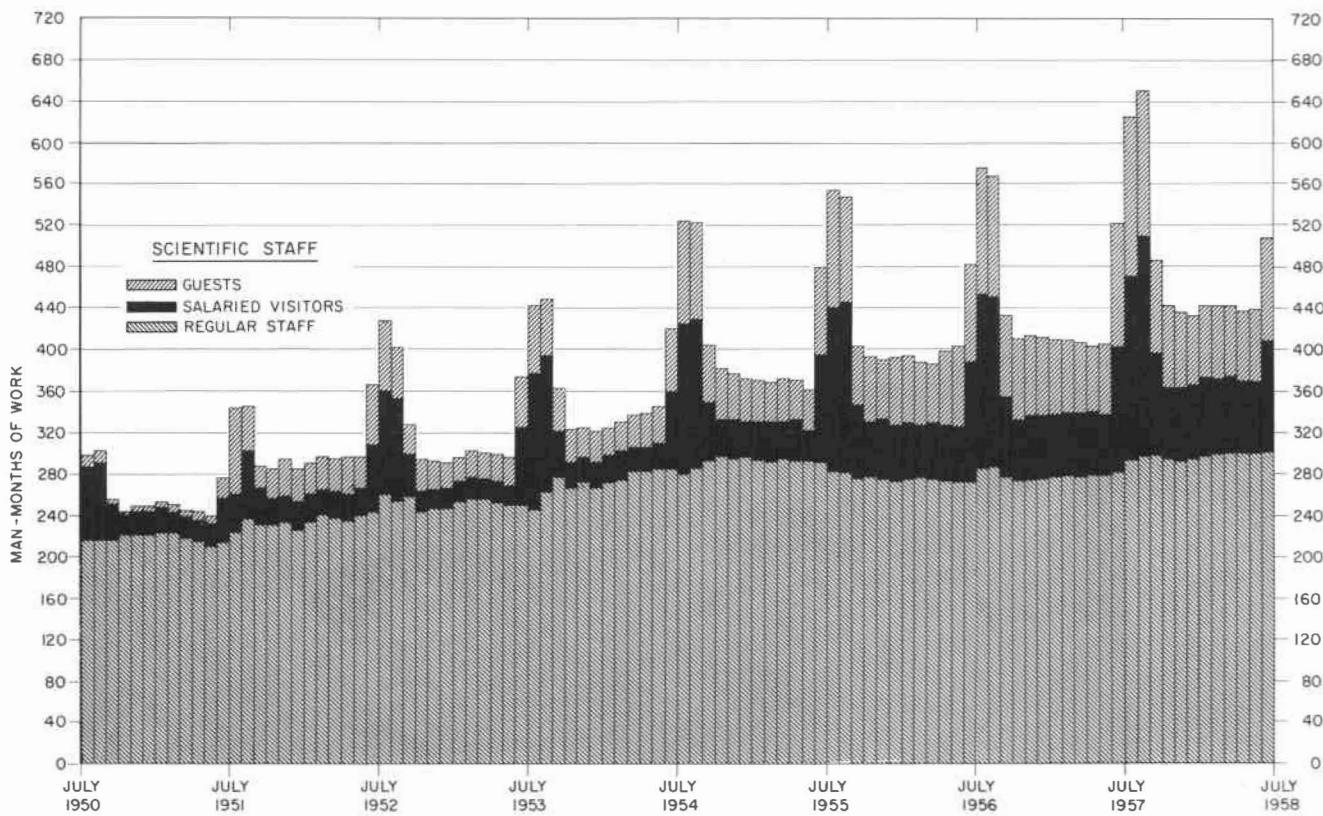


Figure 14.

temporary appointees, research collaborators, and guests. The net increase for the year was one of the largest in several years - 132 as compared with 98 in the preceding fiscal year. The ratio of one scientist to four nonscientists was maintained.

The growth of the scientific staff from July 1950 to July 1958 is shown in Figure 14. The number of man-months of work performed by the scientific staff is plotted for each month. The graph gives, therefore, an indication of scientific services rendered and also represents the average number of scientists working at the Laboratory full time each month. Salaried visitors include scientists and engineers on leave from their own institutions, graduate students doing doctoral investigations while on leave from their graduate schools, post-doctoral research associates on one-year appointments, and temporary appointees whose services are in some measure requested by Brookhaven. Guests include scientists and engineers who participate in the Brookhaven program but receive no remuneration from the Laboratory. The services of consultants are not included in the graph. During fiscal 1958, 14 man-months of service were rendered by these consultants as compared with 17 in the previous year.

For the first time in several years there has been a small but noticeable increase ( $\approx 6\%$ ) in the regular scientific staff as a result of the increasing personnel needs of the Applied Mathematics Division and of such projects as the Liquid Metal Fuel Reactor and the Alternating Gradient Synchrotron. For the 11th successive year the number of scientific visitors who worked at the Laboratory for one month or more has increased; for the salaried and nonsalaried groups the increase is the same,  $\approx 14\%$ .

Foreign scientists participating in research activities at Brookhaven numbered 120 - exactly double the number for the fiscal year ending June 30, 1956. The presence of foreign scientists working cooperatively with others at the Laboratory continues to be stimulating to scientific advancement and mutual understanding.

Arrangements have been made for 76 faculty members from 45 educational institutions to work at the Laboratory during the summer of 1958. About 17 scientists and engineers from other organizations are also expected. The formal summer program for students will bring 41 undergraduates and 20 graduate students from 40 educational institutions. Including 18 Radiological Physics

Fellows and 17 additional graduate students, a total influx of about 200 additional research workers is expected this summer.

### **SUMMER INSTITUTE IN NUCLEAR SCIENCE AND ENGINEERING**

For the second successive year an eight-week Summer Institute in Nuclear Science and Engineering, sponsored by the American Society for Engineering Education and the Atomic Energy Commission, was held at the Laboratory for 30 professors from engineering schools. The objective was to present to the faculty members the basic theory and technology of reactor development, together with pertinent laboratory experiments. The program was similar to that given in the summer of 1956 and described in the previous annual report. About 30 members of the Laboratory's scientific staff contributed services in their respective fields. A reactor symposium to which nine full mornings were devoted at the end of the course served to integrate the material presented earlier. Experts directly engaged in a number of power reactor projects gave the lectures, which were open to all Laboratory staff members. Discussion periods for the Institute members followed the lectures. The interest and response of the group from the engineering schools was again most heartening and more than repaid the Laboratory staff for their efforts in presenting the program.

During the coming summer, four similar institutes are being sponsored by the same two organizations at the University of California (Berkeley), Cornell University, Purdue University, and North Carolina State College (Raleigh). For the Cornell University group the Laboratory has agreed to arrange a program of special experiments, tours, and lectures at Brookhaven during the last ten days of the eight-week course.

### **PROJECT COLOMBO**

Early in 1956, the Laboratory undertook a detailed study to assist the U.S. Government in drawing up plans for the installation, operation, and administration of a regional nuclear research and training center to be located in the Philippines, toward which this country would make a major financial contribution. A study group, known locally as Project Colombo, was formed and a tour

was made of 14 of the Asian countries within the Colombo Plan.

Following the tour, the project staff prepared a report based on its findings. This was distributed by the U.S. Department of State to the member nations of the Colombo Plan for their study and comment, with the suggestion that a working group be established to discuss the Brookhaven report and to formulate specific recommendations to the respective governments. Accordingly, a two-week Asian Nuclear Center Working Group meeting was held in Washington in July 1957. Prior to this meeting, a three-day visit to Brookhaven was arranged for the delegates, to provide them with the opportunity of examining in the atmosphere of a cooperatively managed nuclear research institution the scientific and technical problems involved in their pending discussions. A series of talks and conducted tours covering the program of most direct concern to the Center were given by key members of the Laboratory staff, and less formal meetings were arranged for individual interest groups.

The subsequent meetings in Washington, which were attended by Brookhaven representatives, disclosed that very few Colombo Plan nations were ready to commit themselves to contributing substantially toward the operating costs of the proposed regional center. At the same time, it was evident that most of the Asian member countries would welcome a center where their personnel could receive training and conduct research on a scale not now practicable on a national basis. The U.S. Government therefore decided to study the possibilities of modifying its original proposal of a center to be established within the framework of the Colombo Plan, and in its stead to consider a joint U.S.-Philippine center, far more modest in its initial scope than that proposed originally. In its early stages, this center would be devoted mainly to training, with only a modicum of research activity; and other countries presumably would not be required to provide substantial financial support.

In line with this new approach, the U.S. Atomic Energy Commission was requested to prepare a technical plan for a modified center, to be constructed and operated for several years with an expenditure not to exceed \$10 million. In March 1958 Brookhaven staff from the original Project Colombo collaborated with the AEC's Division of International Affairs in the preparation of the

technical plan requested by the Department of State. This revised plan is currently being reviewed by the U.S. Government.

## GENEVA CONFERENCE

The Second International Conference on the Peaceful Uses of Atomic Energy is to be held at Geneva beginning September 1, 1958. C. Williams is serving as a member of the staff of the Technical Director of the Office of the International Conference. A complete demonstration of the experiment by which the helicity of the neutrino was determined here at Brookhaven has been prepared and will be on exhibition. Among those in charge of this demonstration will be G. Goldhaber, M. Goldhaber, L. Grodzins, and A. Sunyar.

As delegates, the following Brookhaven staff members will present the papers listed below.

J. BIGELEISEN: Temperature Independent Isotope Effects in Chemical Exchange Equilibria Involving Linear Molecules.

V.P. BOND: Mechanism of Induction of Mammary Neoplasm in Rats by Radiation: Relations to Dose and Ovarian Status.

J. CHERNICK: A Review of Resonance Capture in Lumps.

H. J. CURTIS: Radiation-Induced Aging in Mice.

D. J. HUGHES: Neutron Cross Sections of Interest to Reactor Design.

W. L. HUGHES: Recent Advances in the Application of Tritium to Biological Studies.

H. J. C. KOUTS: Physics of Slightly Enriched Normal Water Lattices.

A number of other scientific reports from this Laboratory have been prepared for the Conference but will not be presented in person. These are listed with other publications in Appendix A.

The Laboratory's Public Information Office cooperated with the AEC in the preparation of technical material for the 1958 Geneva Conference. Brookhaven is featured in several films being produced for this conference. The Medical Research Reactor is the subject of one of these films. Another, on particle accelerators, includes considerable footage on the Alternating Gradient Synchrotron and the Cosmotron. Several other short films are being made here, notably one on the labeling of thymidine with tritium. Several

technical exhibits are also in the course of preparation.

## CONFERENCES AND PUBLIC INFORMATION

Thirteen formal conferences were conducted at the Laboratory during the past year:

- 1) Working Group Meeting on Asian Nuclear Center, July 9-12, attended by 40 Asians representing 15 nations and 17 United States participants;
- 2) Sixth Annual Naval Reserve Nuclear Sciences Seminar, on Nuclear Reactors and Power, Sept. 8-21, attended by 45 officers;
- 3) Nuclear Shielding Research Review Meeting, sponsored by Aeronautical Research Laboratory, Sept. 11-12, attended by 15 scientists;
- 4) AEC Bio-Med Program Directors Meeting, Oct. 7-8, attended by 34 AEC and contractors' employees;
- 5) Materials Management Workshop, sponsored by the AEC, Oct. 8-11, attended by 13 AEC and contractors' employees;
- 6) AEC Contractors Personnel Conference, Oct. 24, attended by 22 individuals;
- 7) Conference on University Relations, Nov. 1-2, attended by 12 administrative officers from the national laboratories;
- 8) Molecular Beam Conference, Nov. 1-2, attended by 90 individuals representing 29 institutions;
- 9) French-American Conference on Graphite Reactors, Nov. 12-15, attended by 36 persons, 10 from France, 2 from Sweden, and 24 from the United States;
- 10) A classified symposium on Thorium and  $U^{233}$ , sponsored by the AEC, Jan. 9-10, participated in by 21 AEC and 62 contractors' representatives;
- 11) Second Conclave on Nuclear Energy in Medicine, jointly sponsored by the Division of Biology and Medicine of the AEC and BNL, Feb. 6-7, attended by 78 pathologists;
- 12) Conference on Bubble Chamber Scanning and Data Handling Devices, May 26-27, attended by 67 scientists representing 30 institutions; and
- 13) The Eleventh Annual Brookhaven Biology Symposium, on the Photochemical Apparatus, Its Structure and Function, June

16-18, attended by 139 scientists representing 48 institutions.

During the report period, 1946 individuals representing 57 professional and technical groups in this country paid one-day visits to the Laboratory to view its facilities and to hear about its research program.

On October 11 the fourth annual Student Visitors' Day was attended by 5677 students, representing about 100 schools and colleges. On October 12, the eighth annual Visitors' Day, 3632 persons toured the Laboratory facilities. The total of 9309 represents an increase of 1409 over the previous year.

The AEC, in commemoration of Thomas Alva Edison's birthday, asked its contractors to invite outstanding high school science students and editors of school papers to their installations. Brookhaven invited 42 high schools in Suffolk County to participate in this event. Of these 26 schools responded, and the 248 selected students who visited the Laboratory on that day participated in a panel discussion on "What Makes a Scientist" and then visited the major facilities.

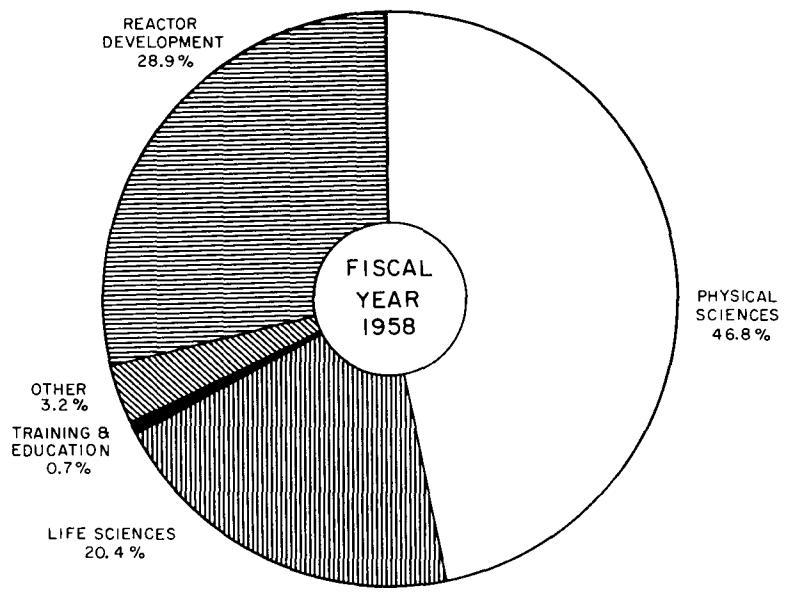
In addition, the Laboratory has been host for one or more days during the year to nearly 1000 professional, governmental, or industrial representatives of other countries.

The normal activities of the Public Information Office have continued to increase, mainly because of the heightened interest shown by the general public in the nuclear sciences. Staff members have responded to 52 requests to address lay and semiprofessional audiences in the vicinity of the Laboratory. Requests for information and literature from schools and from individual students reflect the considerable efforts being made by educational authorities in this country to stimulate the adoption of careers in science and engineering. Loans of technical motion pictures from the Brookhaven film library reached an all-time high during the past winter months.

## ADMINISTRATION

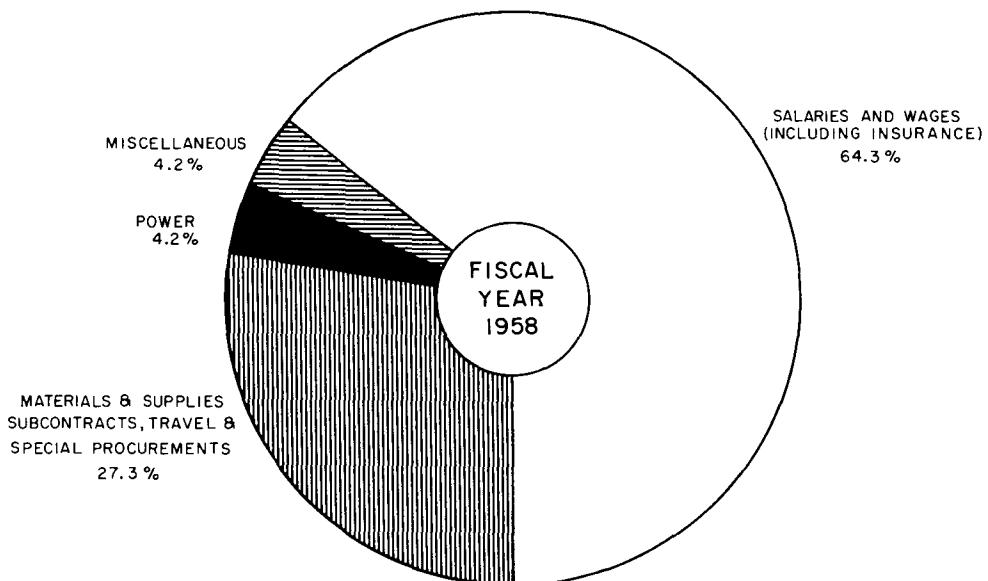
The organization of the Laboratory as of July 1958 is given in Figure 1. The most important changes during the past year are as follows.

Joseph S. Washburne, Jr., was appointed Personnel Manager effective April 30, 1958; he succeeds Edward A. Macy who has resigned to



OPERATING COST DISTRIBUTION BY PROGRAMS

FISCAL YEAR	PHYSICAL SCIENCES	REACTOR DEVELOPMENT	LIFE SCIENCES	TRAINING & EDUCATION	OTHER (NET)	TOTAL COST
1958	8,281,717	5,114,706	3,609,035	124,116	577,726	17,707,300
1957	7,004,220	3,657,329	3,021,186	54,069	684,133	14,420,937
1956	6,201,158	2,543,633	2,637,958	61,183	420,151	11,864,083



MAJOR CATEGORIES OF OPERATING EXPENDITURES

FISCAL YEAR	SALARIES AND WAGES	MATERIALS & SUPPLIES SUBCONTRACTS, TRAVEL & SPECIAL PROCUREMENTS	POWER	MISCELLANEOUS (NET)	OPERATING TOTAL
1958	11,381,555	4,841,907	742,006	741,832	17,707,300
1957	9,885,738	3,428,550	752,879	353,770	14,420,937
1956	8,726,773	2,189,998	711,590	235,722	11,864,083

Figure 15.

accept another position. Mr. Washburne has been a member of the Laboratory staff and of the Personnel Division for the past eleven years.

George W. Maxey was appointed Purchasing Agent effective April 30, 1958. Mr. Maxey succeeds Charles H. Keenan, Purchasing Agent for the past several years, who has resigned to accept another position. Mr. Maxey was most recently in the Director's Office where he worked on organization and methods.

## FINANCE

Excluding the direct costs of the Alternating Gradient Synchrotron and other plant and equipment items, organizational costs for fiscal 1958 totaled \$17,707,300, an increase over the previous year of \$3,286,363 (+22.8%). The Laboratory's research programs directly supported by the AEC showed a total increase of \$3,392,770 (+24.7%) over the previous year, while work for others declined \$106,407 (-15.7%).

During the year the AEC established a new program classification entitled Training and Education, to which were charged certain BNL activities previously supported by the Divisions of Reactor Development and Biology and Medicine, and some new activities. The Laboratory's AEC-supported research was financed, therefore, by four Divisions, Reactor Development, Research, Biology and Medicine, and Training and Education. Total costs of research in these four programs were \$17,129,574, an increase of nearly 25% over the previous fiscal year. (See Figure 15.)

Of the total cost increase of \$3,392,770 in AEC-supported research, \$2,073,228 (61.1%) occurred in the direct costs of research. Nuclear Engineering activities, particularly those connected with the LMFR, accounted for \$1,040,707 (50.2%) of the direct cost increase. Expenditures for LMFR development subcontracts and special procurement were responsible for \$755,489 (72.6%) of the total Nuclear Engineering increase. Similarly, in Physics and Chemistry the major portion of the direct research cost increase of \$663,195 (32.0% of the Laboratory's total in this category) was attributable to special procurements. These increased special procurement costs resulted from completion of acquisition of the enriched fuel elements for reloading the reactor and the beginning of delivery of the new coils for the Cosmotron repair work.

The direct cost increase in Biology and Medicine is attributable largely to higher costs of doing business. There was essentially no change in total staff during the year. The Laboratory again participated in the Government's continuing Marshall Islands studies. At the request of the AEC and other government agencies, the Laboratory now includes these studies in its research programs. The Laboratory also was host this year for the AEC's Medical Conclave.

Training and Education, the new AEC activity mentioned above, includes at BNL the Special Training and Summer Institute activities previously supported by other research programs. In addition it includes the participation by the Laboratory in the AEC's interests in world-wide co-operation to promote the peaceful uses of atomic

Table 2  
Capital Equipment Expenditures and Commitments  
(Including Charges from Organizational Units, See Table 1)

	FY 1958		FY 1957		FY 1956	
	\$	%	\$	%	\$	%
Scientific & hospital	1,552,880	74.6	1,066,359	79.0	582,430	80.5
Automotive & heavy mobile	206,119	9.9	98,725	7.3	40,894	5.6
Office machines & furniture	87,267	4.2	72,956	5.4	35,348	4.9
Shop equipment	149,468	7.2	70,003	5.2	49,843	6.9
Miscellaneous	85,480	4.1	41,630	3.1	15,298	2.1
Expenditures & commitments, Total	2,081,214	100.0	1,349,673	100.0	723,813	100.0
Proceeds from sales	(27,997)		(16,107)		(18,773)	
Expenditures & commitments, Net	2,053,217		1,333,566		705,040	

Table 3

Costs Incurred for Fixed Assets  
(Including Charges from Organizational Units, See Table 1)

	FY 1958			FY 1957			FY 1956		
	Man-years			Man-years			Man-years		
	Costs	Sci.	Others	Costs	Sci.	Others	Costs	Sci.	Others
<b>ALTERNATING GRADIENT SYNCHROTRON</b>									
Direct									
Salaries, wages, insurance	748,473	30.5	77.0	585,480	26.5	60.5	492,460	23.5	47.5
Materials, construction, etc.	4,897,065			3,773,723			1,871,898		
Subtotal direct	5,645,538			4,359,203			2,364,358		
Charges from organizational units	162,269			125,031			117,697		
Total	5,807,807			4,484,234			2,482,055		
<b>OTHER, INCLUDING MEDICAL RESEARCH CENTER REACTOR</b>									
Direct									
Salaries, wages, insurance	98,035	4.0	8.0	39,273	2.0	3.0	10,565	1.0	0.5
Materials, construction, etc.	1,924,177								
Subtotal direct	2,022,212			1,556,165			948,125		
Charges from organizational units	90,932			93,618			69,201		
Total	2,113,145			1,649,783			1,017,326		

energy. Laboratory costs this year were \$70,047 (129.5%) greater than in fiscal year 1957.

Table 1, on the reverse side of the organization chart (Figure 1), shows in detail the operating expenditures on a broad organizational basis. While labor costs continued to be the largest single item of expense, the increase in costs for development subcontracts and special procurements, especially in connection with the LMFR, reduced the proportion expended for salaries and wages to 64.3%. Last year the figure was 68.5%, and it has been as

high as 75.0%. The charts and tables in Figure 15 present the breakdown of costs by major categories of expenditures, as well as by major programs.

Table 2 gives the expenditures and commitments for capital equipment. A summary of expenditures for fixed assets (plant and equipment) is shown in Table 3.

Upward economic pressures continued during the year. Salary and wage rates advanced  $\approx 7\%$ , and increases in costs of materials and services averaged  $\approx 5\%$ .

BROOKHAVEN NATIONAL LABORATORY  
BALANCE SHEET

	June 30, 1958	June 30, 1957
<b>ASSETS</b>		
Cash	\$ 465,957	\$ 777,830
Accounts receivable	101,501	128,065
Inventories	476,027	372,783
Advances and prepaid items	245,868	190,184
Deposits	308,871	18,938
Fixed assets (less reserves of \$23,646,474 at June 30, 1958, and \$23,993,640 at June 30, 1957)	36,670,859	34,614,893
Construction in progress	15,701,555	9,201,338
Total assets	\$53,970,638	\$45,304,031
<b>LIABILITIES</b>		
Accounts payable	\$ 2,138,729	\$ 1,499,043
Accrued payroll	126,577	88,757
Atomic Energy Commission	51,705,332	43,716,231
Total liabilities	\$53,970,638	\$45,304,031

PHYSICAL  
SCIENCES  
AND  
ENGINEERING



# Physics

The research program in physics continues to consist of experimental and theoretical studies concerned with the structure and fundamental properties of matter. Through observations of the interactions of charged particles, neutral particles, and radiation with matter, information is sought which will provide a better understanding of the complex character of the structure of matter. Some of these studies deal with interactions involving individual atoms, atomic nuclei, or nucleons, and others with the characteristics of conglomerates of atoms and molecules in bulk matter. Various features of the structure and properties of matter can be most effectively studied with particles or radiation of a particular energy or range of energies. To this end the Cosmotron, 60-in. cyclotron, Van de Graaff accelerator, 18-in. cyclotron, and reactor at Brookhaven provide a wide range of energies and a wide diversity of particles and radiation for experimental investigations. The experimental researches reported here center on one or more of these major facilities and will be described under the categories of high energy physics, nuclear structure, neutron physics, atomic and molecular physics, and solid state physics. The theoretical scientists conduct their investigations in close association with the experimental research, and they provide stimuli to the search for new results and new approaches to the problems.

A number of visiting and guest scientists, on leave from other institutions in this country and abroad, came here this past year to carry out research at one or another of the Brookhaven facilities. These scientists are attracted not only by the availability of these facilities but frequently by the opportunity to collaborate with members of the Brookhaven staff. Their research augments the scientific effort, and their association with the staff provides the cross fertilization of ideas and experience so important to the physics research program.

## HIGH ENERGY PHYSICS

The breakdown of the Cosmotron restricted experimental work during the last year to a period of

only a few months. However, a considerable amount of data was collected during this time, which has served to expand our knowledge of the properties of the so-called strange particles, the heavy mesons and hyperons. In particular, data have been collected concerning the reactions  $\pi^- + p \rightarrow \Lambda^0 + K^0$ ;  $\pi^- + p \rightarrow \Sigma + K$ ; and  $\pi^+ + p \rightarrow \Sigma^+ + K^+$ . Analyses of these measurements have established to a high degree of probability that the spins of the  $\Sigma$  and  $\Lambda$  hyperons are  $\frac{1}{2}$  and the spin of the  $K$ -meson is zero.

The investigation which led to the discovery that parity is not always conserved in nature was mainly instigated by evidence that the  $K$ -meson violated parity conservation in its decay. With the exception of the not wholly conclusive interpretations concerning evidence of the decay characteristics of this particle, parity nonconservation had been observed only in reactions in which a neutrino was emitted. The analysis of the  $\pi^- + p \rightarrow \Lambda^0 + K^0$  reaction has now shown that the decay of the  $\Lambda^0$  also violates parity. The pertinent observation concerns the fact that for an incident pion beam traveling north,  $\Lambda^0$ -particles produced traveling east prefer to decay so that protons are emitted up, rather than down. It can be shown that these relationships between the directions involved will be changed if the experimental results are observed reflected in a mirror. This is then an indication that parity is not conserved in the decay of the  $\Lambda^0$ . The effect is quite large, which indicates that about half of the decay intensity has even parity and about half has odd parity. A further analysis of the results indicates that the reaction is not invariant under charge conjugation; that is, the relationship of the spatial vectors will be different if particles are replaced by antiparticles. However, it appears possible that the product of the charge conjugation number and parity quantum number may be conserved. In other words, if the interaction is observed in a mirror and particles are replaced by antiparticles, the relationships between the observed quantities will remain the same, or be invariant under the two transformations.

Measurements of the properties of the reaction  $\pi^+ + p \rightarrow \Sigma^+ + K^+$  near threshold are nearly com-

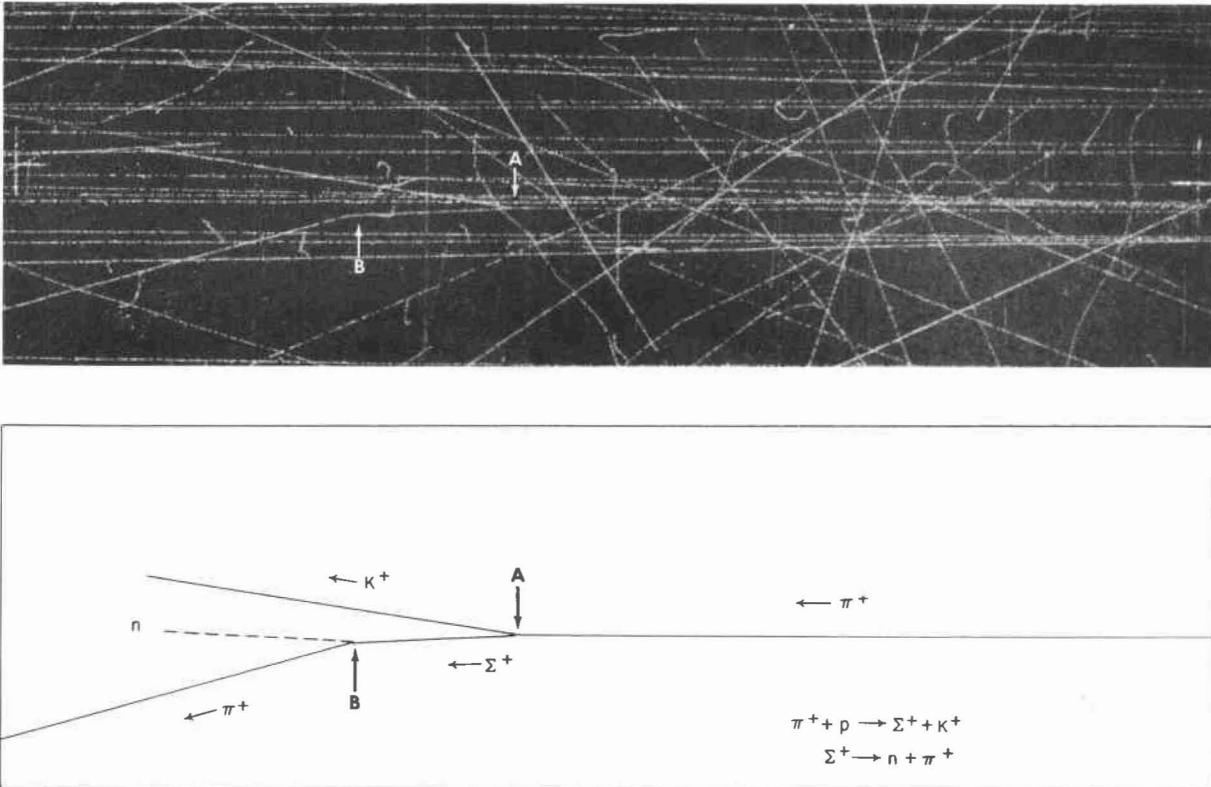


Figure 1. Bubble chamber picture of associated production. The sketch serves to clarify the photograph. A positive pion interacts with a proton at point *A* to produce a positive  $K$ -meson in association with a positive  $\Sigma$ -hyperon, which very shortly, at point *B*, disintegrates into a positive pion and a neutron. The path of the neutron is not seen in the photograph because the neutron is uncharged and does not produce bubbles along its path.

pleted. These results suggest with poor statistical accuracy that the cross section for the production of  $\Sigma^+$ -particles by  $\pi^+$ -mesons incident on protons may not be very different from the cross section for the production of  $\Sigma^0$ -mesons by  $\pi^-$ -mesons near threshold. The  $\pi^+ + p \rightarrow \Sigma^+ + K^+$  reaction was observed by passing  $\pi^+$ -mesons through a hydrogen bubble chamber and a propane bubble chamber. The hydrogen bubble chamber pictures have been analyzed to determine the  $\pi^+, p$  elastic differential scattering cross section and the  $\pi^+, p$  inelastic scattering cross section. The elastic differential scattering cross section is qualitatively quite different from the  $\pi^-, p$  elastic differential scattering cross section measured at the same  $\pi$ -meson energy. In particular the shape and width of the elastic scattering peak in the forward direction are quite different for  $\pi^-, p$  scattering than for  $\pi^+, p$  scattering. Measurements of the  $\pi^-, p$  elastic scattering differential cross sections at various energies between

500 and 900 Mev have been made by use of propane and hydrogen bubble chambers. The results show qualitative features which vary continuously with energy throughout this region. Properties of the elastic scattering cross sections are of particular interest in this region because of the probable existence of nucleon isobars at this energy.

Extensive measurements have been made on the production of  $K$ -mesons in light nuclei and the interactions of  $K$ -mesons with emulsion nuclei.

Yields of magnetically selected  $K^+$  and  $K^-$ -mesons produced in proton bombardments of Be (and Pb) and detected in nuclear emulsions, were measured at different proton energies. The differential production cross section for 300-Mev/c  $K^+$ -particles, ejected from a Be nucleus at  $\approx 0^\circ$  to a beam of 3-Bev protons, is  $4 \times 10^{-6}$  mb per steradian Mev/c. The yields of both species fall sharply with diminishing proton energy, the  $K^-$  yield to a greater extent; the yield ratio  $K^+/K^-$  is about 30 at 3 Bev and

about 300 at 2 Bev, and at 1.7 Bev the  $K^-$  yield is undetectable, whereas the  $K^+$  yield is still appreciable.

The principle of conservation of strangeness requires, in nucleon-nucleon collisions, a different reaction for production of  $K^+$  than for production of  $K^-$ -mesons. The phase-space yields of the theoretically permissible reactions, with due allowance for the nucleon momenta within the nucleus, were computed; the effects of intranuclear  $K$  scattering upon yield were also estimated. The energy dependences of the resulting yield curves are roughly consistent with the above-mentioned observations.

Observations of nuclear scattering of  $K^+$ -mesons in emulsion nuclei, previously reported for  $K$  energies below 150 Mev, have been extended to 300 Mev. About  $\frac{1}{3}$  of all  $K$  scatterings are inelastic. The inelastic scattering cross section of these mesons increases with  $K$  energy; the cross section ratio  $\sigma_{300}/\sigma_{150} \approx 1.5$ . Furthermore, the charge-exchange fraction (the fraction of inelastic collisions in which  $K^+ \rightarrow K^0$ ) rises also with energy, about doubling between 150 and 300 Mev, where  $\frac{1}{3}$  of the inelastic collisions are charge-exchange.

By means of an optical model, the  $K$  nuclear scattering was analyzed, and, from this analysis and the measurements,  $K$ -nucleon cross sections at various  $K$  energies were computed. These too show a rise with  $K$  energy in the 150 to 300 Mev region. It is interesting to note that in the 140 to 218-Mev interval, this  $K^+$ -nucleon cross section has the same value as the  $K^+ - p$  cross section, measured, from  $K-p$  collisions identified in these observations, to be  $15 \pm 6$  mb.

Measurements of the inelastic scattering of  $\pi^-$ -mesons by protons are in progress. Interactions taking place in a hydrogen bubble chamber are being analyzed.

Assembly of a  $20 \times 10 \times 9$ -in. hydrogen bubble chamber is now almost complete. The 17,000-gauss magnet and practically all chamber components have been tested, and performance has been found satisfactory. A semiautomatic measuring projector is under construction for analysis of the photographs expected when the Cosmotron resumes operation. An LGP-30 automatic computer has been purchased, and programs are being written for evaluation of measurements.

The small 6-in. chamber constructed as a working model for the 20-in. chamber has been exposed to Cosmotron radiation of varying energies in order to explore the bubble density as a func-

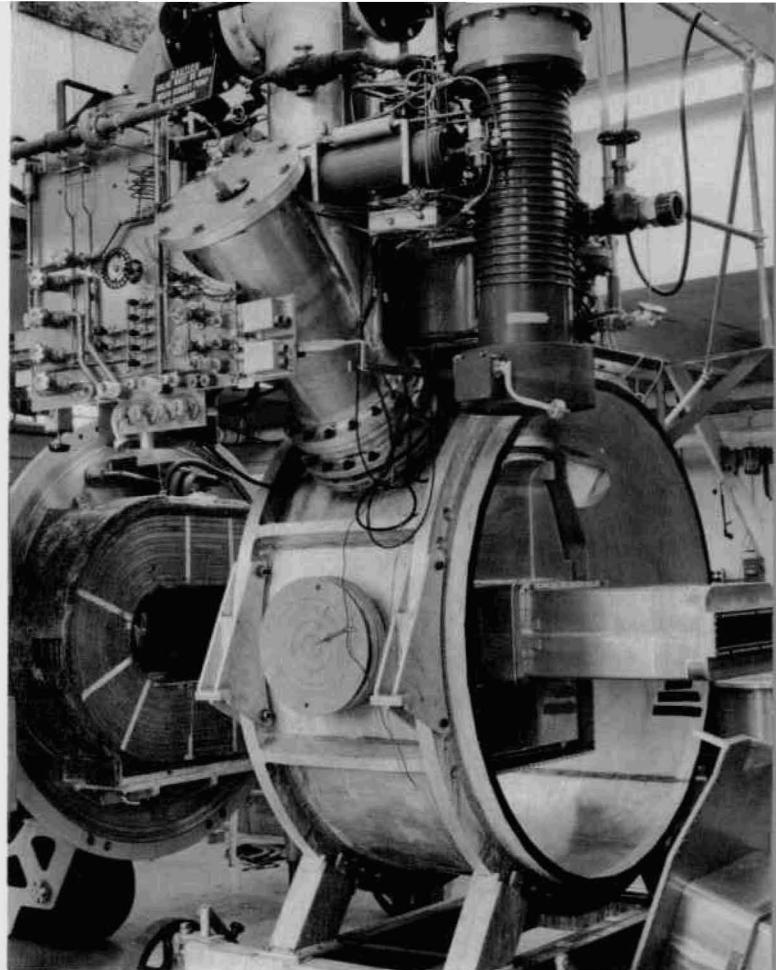


Figure 2. In the above picture major parts of a  $20 \times 10 \times 9$ -in. liquid hydrogen bubble chamber are shown. The chamber itself is located at the center of the  $5\frac{1}{2}$ -ft-diameter circular aluminum vacuum tank. Towards the right protrudes a heat shield. At the top can be seen the diffusion pumps which produce a high vacuum in the tank, and various controls. Behind the tank (left side of picture) appears one half of the magnet consisting of a copper coil insulated in epoxy plastic, and a large steel yoke. Also visible at lower right is one of the end covers for the vacuum tank. The chamber will be filled with liquid hydrogen, which when expanded will be superheated by a mechanism at the top of the vacuum tank, so that tracks of charged particles from the Cosmotron will become visible when illuminated through the slot at the right side of the heat shield and photographed from the other side.

tion of velocity. The expected dependence on the inverse of the square of the velocity was not confirmed. Further experiments on this question bearing heavily on data evaluation will be performed with the 20-in. chamber. The small chamber is also being used for an investigation of the operating characteristics of liquid deuterium.

Preliminary discussions and studies have been carried out on construction of a much larger hydrogen bubble chamber measuring  $80 \times 25 \times 20$  in.

for use at the Alternating Gradient Synchrotron. This size is dictated by the high energies encountered there, which necessitate long tracks and long decay paths within the chamber for effective measurements.

Theoretical work on particle physics at high energies has continued. The properties of strange particles have naturally been the subject of investigation. In particular the possible similarity or lack of similarity in the internal space-structure of the  $\Lambda^0$  and  $\Sigma^0$  particles has been examined. When a  $\Sigma^0$  decays into a  $\Lambda^0$  by emitting a  $\gamma$ -ray, the energy of the  $\gamma$ -ray can occasionally be converted into an electron-positron pair. The probability of conversion has been calculated on the alternative assumptions that the parent and daughter particles have or do not have similar space-structure (i.e., the same parity). A small but measurable difference is predicted.

Various hypotheses about the interaction of  $K$ -mesons with pions and nucleons have been examined in connection with experiments on the scattering of  $K$ -mesons by nuclei.

The formal theory of the decay of, and reactions between, polarized particles is being examined by a new method which seems especially convenient

in the extreme relativistic region. The method is applicable to particles of arbitrary spin, and it is planned to apply it to a discussion of the reactions in which strange particles are produced.

### NUCLEAR STRUCTURE

Researches in this field may be divided into two broad classifications: those involving the study of the characteristics of unstable nuclei produced by nuclear interactions, and those concerned with the instantaneous products of a nuclear reaction. The term instantaneous is used here in the relative sense and applies to those reaction products which follow the interaction of a particle or radiation with a nucleus in a time too short to be readily resolved with existing techniques. Observations of the ways in which unstable nuclei are produced and transform into stable nuclei (radioactive decay) give information about the characteristics of both the original unstable nucleus and the final stable nucleus. The researches are designed to obtain data which can be interpreted in terms of the identity and energy of the particles or radiation emitted by unstable nuclei. The analysis of these data and the study of the systematics of the decay

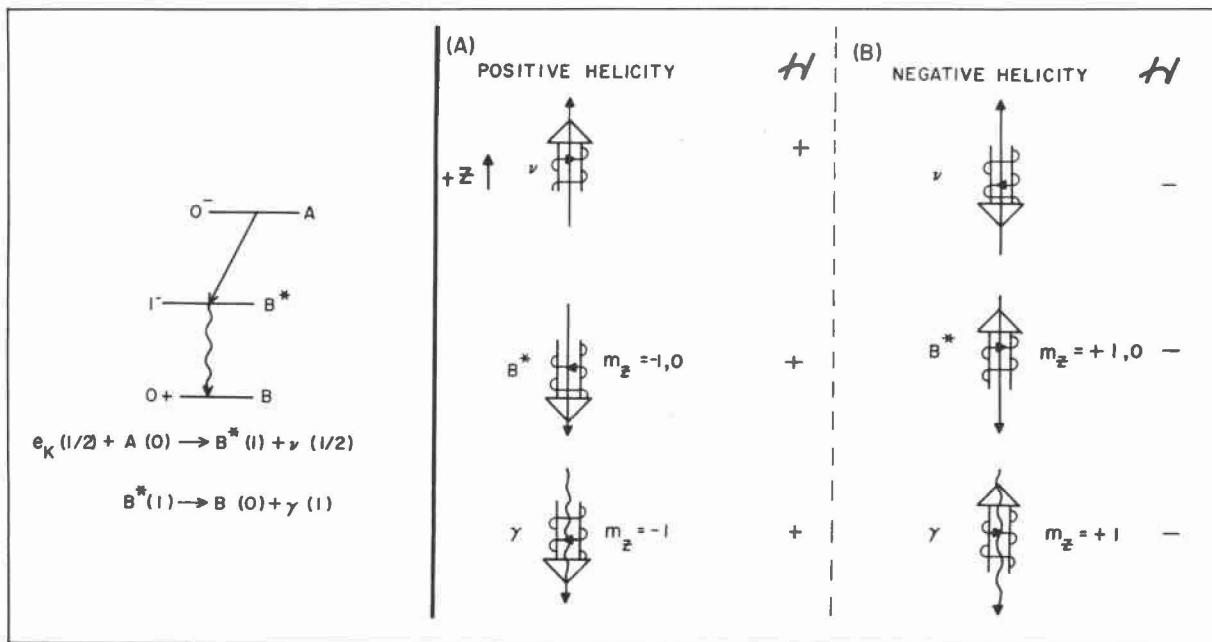


Figure 3. Schematic drawing showing that the helicity of the gamma-ray is the same as that of the neutrino if they are emitted in opposite directions. A. Neutrino has positive helicity and gamma-ray has positive helicity. B. Neutrino has negative helicity and gamma-ray has negative helicity.

process yield information as to the energy level structure and properties of atomic nuclei.

The researches dealing with the instantaneous products of a nuclear reaction also yield information on the energy levels in nuclei. In these cases the identity and energy of the interacting particles or radiation, together with the identity and energy of the products of the reaction, can be interpreted in terms of the energy level structure of nuclei. The ways in which these nuclear levels may be excited and the systematics of the level structure in isotopes consisting of different numbers of protons and neutrons are the basis for the formulation of ideas as to the fundamental structure of matter.

The problems raised by the introduction of the principle that parity is not conserved in some types of nuclear interactions, as discussed in last year's report, continue to be the basis of a number of researches. Since this principle has far-reaching implications in the theoretical considerations of nuclear structure, it is important that it be tested by every possible means. One of the consequences of the nonconservation of parity in beta decay is that the spin vector of the neutrino is either parallel or antiparallel to its direction of motion, depending upon the type of interaction which describes the beta decay. The interaction is tensor or scalar if the spin and momentum vectors are in the same direction, i.e., they are said to be parallel; and the interaction is vector or axial-vector if the spin and momentum are oppositely directed, i.e., antiparallel.

An experiment has been performed which shows that the spin and momentum vectors for a neutrino are antiparallel, that is, a hypothetical spot on the side of a neutrino will move counter-clockwise when the neutrino is moving away from an observer. The sense of the rotation, corresponding to the spin of a particle, relative to the direction of its linear momentum has come to be referred to as the helicity of the particle. If the sense of rotation is clockwise when a particle is moving away from the point of observation, the helicity is said to be positive. (This is the relation which exists between rotation and forward motion for a right-handed screw.) Negative helicity is the converse, i.e., it corresponds to a left-handed screw.

The helicity of the neutrino was determined by measuring the circular polarization of the gamma-rays emitted in the direction opposite to that of the neutrino in a radioactive decay process. The principal details involved in this experiment may be

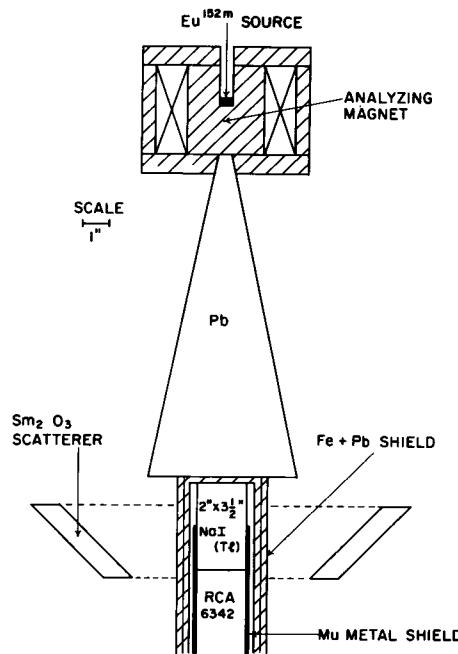


Figure 4. Schematic diagram of the arrangement used in the experiment performed to determine the helicity of the neutrino.

described by referring to the decay schemes represented in the left portion of Figure 3. The nucleus  $A$  (spin 0) decays by capturing an orbital electron (spin  $1/2$ ), and simultaneously emitting a neutrino, to an excited state of nucleus  $B^*$  (spin 1). The conservation of linear momentum requires that the recoiling nucleus  $B^*$  and the neutrino move in opposite directions. Also the conservation of angular momentum requires that the spin (1) of  $B^*$  be opposite to the neutrino spin ( $1/2$ ), since the sum of the initial spins – of the initial nucleus  $A$  and the electron – is  $1/2$ . Thus the helicity of the recoiling nucleus  $B^*$  must be the same as that of the neutrino. The two possibilities corresponding to positive and negative neutrino helicity are depicted in parts  $A$  and  $B$  of Figure 3. The choice between these two possibilities is made by observing the helicity of the gamma-rays emitted when the nucleus  $B^*$  decays to its ground state  $B$  (spin 0). In this process the gamma-ray carries away the one unit of spin. The conservation of angular momentum requires preservation of spin direction. Therefore, if the gamma-ray is emitted in the direction of the recoiling nucleus  $B^*$ , as indicated in Figure 3, it will have the helicity of  $B^*$  and hence of the neutrino. The measurement of the helicity of the neutrino has thus been reduced to the obser-

vation of the circular polarization of the gamma-rays emitted opposite to the neutrino.

The experimental arrangement is shown in Figure 4. The gamma-rays following the decay of  $\text{Eu}^{152m}$  which have been given extra energy by being emitted forward from the recoiling nucleus  $\text{Sm}^{152*}$  (this is significant only if the lifetime of the  $B^*$  state is short compared with the duration of the recoil motion, as is the case for  $\text{Sm}^{152*}$ ) can cause resonance in the samarium scatterer which is placed in the form of a ring around a scintillation detector. The gamma-rays emitted in any other direction rarely have enough energy for resonant scattering. The detector is shielded from the direct radiation from the source and hence detects principally the resonant scattered gamma-rays. The circular polarization of these gamma-rays is observed by utilizing the Compton scattering in magnetically saturated iron. The change in the number of resonant scattered gamma-rays as the direction of magnetization of the iron is reversed gives a measure of the helicity of the gamma-rays and hence, as described above, also determines the helicity of the neutrino. The result of this measurement is that the helicity of the neutrino is negative, corresponding to an axial-vector type of interaction in this decay process (schematically represented in part *B* of Figure 3).

The experiment on the helicity of the neutrino has been the starting point for various theoretical investigations. In particular the theory of angular correlations in forbidden beta decay has been worked out in detail. An application of these results is being made to the famous old problem of the RaE spectrum. It appears at present that the circular polarization of the beta-rays of RaE can be predicted correctly if the matrix elements of the decay are chosen in such a way as to fit the shape of the continuous spectrum.

One result of the many experimental and theoretical investigations performed since the discovery that parity is not conserved in weak interactions was the demonstration that previous beta-recoil correlation experiments are not self-consistent. In the case of  $\text{He}^6$ , the original correlation data together with the assumption that  $\text{He}^6$  has zero spin lead to a disagreement with the theory of a universal Fermi interaction. To investigate this difficulty, experiments are in progress under the Columbia-BNL cooperative program to measure both the spin and beta-recoil correlation for  $\text{He}^6$ . A system to produce the gas by the reaction

$\text{Be}^9(n,\alpha)\text{He}^6$  has been constructed and installed in the BNL reactor. The apparatus for the spin measurement is essentially of the Stern-Gerlach type. A beam of  $\text{He}^6$  atoms from a narrow slit traverses an inhomogeneous magnetic field, then passes through a long narrow channel, and enters a cavity defined by thin aluminum walls. The beta active atoms, which have a very small probability of leaving the cavity through the channel before decaying, are detected by a scintillation counter. If the  $\text{He}^6$  nuclei have an appreciable magnetic moment, the beam will be deflected from the channel entrance when the magnet is energized. It is estimated that a nuclear moment of 0.1 nuclear Bohr magnetons will give a change in counting rate of 7%. If the upper limit placed on the moment by this method is very small, it would be very probable that the spin of  $\text{He}^6$  is zero. The design and construction of the apparatus for the  $\text{He}^6$  beta-recoil experiment have also been started. The momentum spectra of the recoil  $\text{Li}^6$  ions will be measured by a magnetic analyzer, and the energy of the beta-particle detected in coincidence with the recoil with a beta scintillation spectrometer.

Various electromagnetic effects connected with weak interactions and lack of parity conservation have also been the subject of theoretical investigations. In particular, the consequences of the assumption that some elementary particles might have an electric dipole moment have been examined, and upper limits have been placed on such dipole moments.

In the investigations of the beta-decay interaction, under the Columbia-BNL cooperative program, the study of a special group of mirror transitions which have nuclei with a "doubly closed shell  $\pm$  one nucleon" configuration has been completed. Precise data on the  $f\tau$  values of these transitions together with the transitions with spin change  $0 \rightarrow 0$  are important for determining  $(C_{GT}^2/C_F^2)$ , the ratio of the squares of the Gamow-Teller to Fermi coupling constants in the beta interaction. The last member of this group investigated was  $\text{Ca}^{39}$ . This isotope was produced at the BNL 60-in. cyclotron by the reaction  $\text{K}^{39}(p,n)\text{Ca}^{39}$ . The beta spectrum, measured with a thin-lens spectrometer, had a maximum energy of  $5.49 \pm 0.03$  Mev; and the activity decayed with a  $0.88 \pm 0.01$ -sec half-life. No gamma-rays were observed. The  $f\tau$  value for  $\text{Ca}^{39}$  calculated from these results is  $4325 \pm 125$  sec.

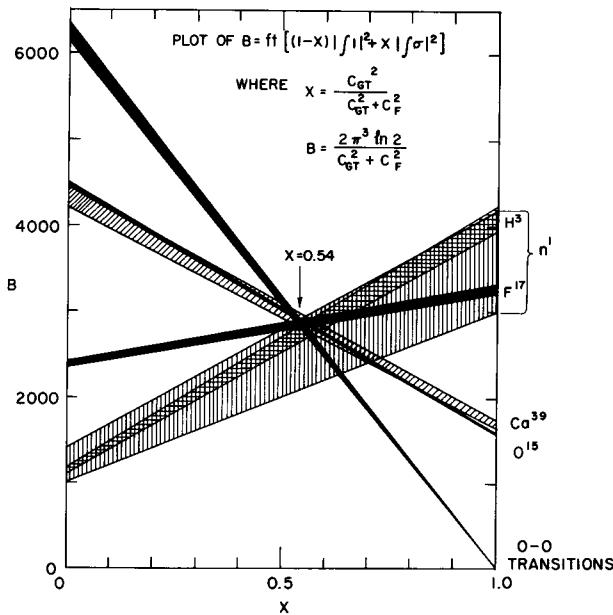


Figure 5.  $B$ - $x$  diagram illustrating the agreement of the “doubly closed shell  $\pm$  one nucleon” mirror beta transitions and the  $0 \rightarrow 0$  spin change transitions with a unique value for the Gamow-Teller and Fermi coupling constants.

The comparison of the  $ft$  values of the “doubly closed shell  $\pm$  one nucleon” and the  $0 \rightarrow 0$  spin-change transitions is shown in Figure 5. The Gamow-Teller matrix elements for this comparison were calculated from the measured magnetic moments of the daughter nuclei. The “lines” for the nuclei identified in the diagram are expected theoretically to intersect at a unique point. Because the experimental  $ft$  values used in this comparison have finite errors, the “lines” are represented by shaded bars. The agreement of all the data is excellent, as can be seen by the intersection of all the bars on the diagram at  $x = 0.54$ . A weighted fit for all the transitions gives a value for  $(C_{GT}^2/C_F^2)$  of  $1.18 \pm 0.07$ .

From the studies of the radioactive decay process it has been shown that the level schemes of the even osmium ( $Z = 76$ ) nuclei form the interesting transition region between the deformed rare earth, Hf, and W isotopes ( $Z \leq 74$ ), which are characterized by rotational bands, and the near-harmonic Pt ( $Z = 78$ ) nuclei whose levels display a near-harmonic pattern. This region has been studied further. Figures 6 and 7 illustrate the nature of this transition. Figure 6 presents the energies of the first excited states of a number of even-even nuclei.

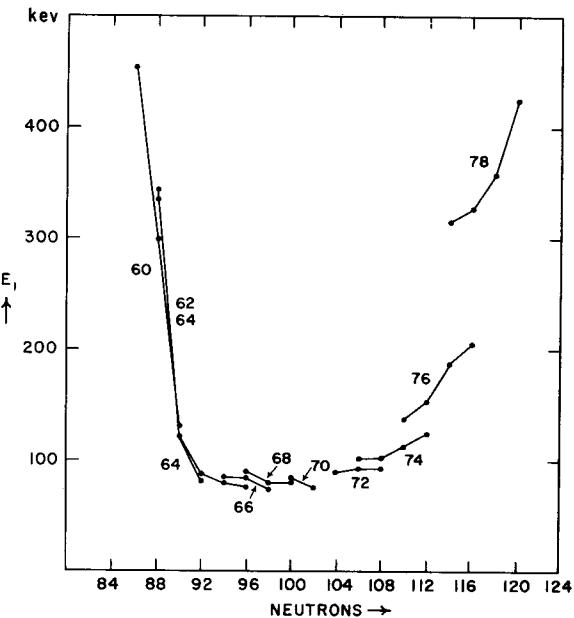


Figure 6. Energies of first excited states of even-even nuclei for  $146 \leq A \leq 198$ . The lines connect points for isotopes of the same element and are labeled with their atomic number. The even osmium ( $Z = 76$ ) nuclei are believed to form the transition region between the deformed rare earth nuclei, characterized by rotational bands, and the near-harmonic nuclei, whose levels display a near-harmonic pattern.

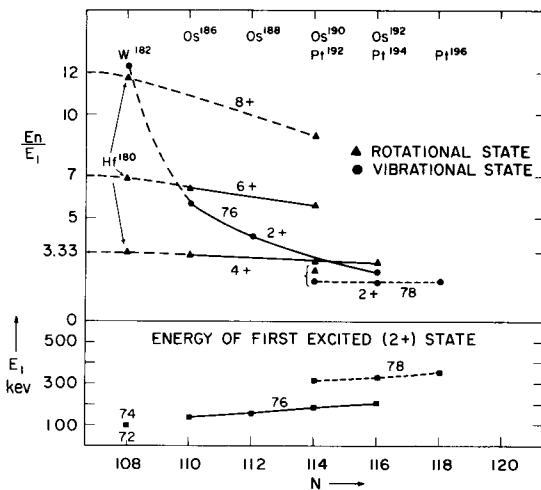


Figure 7. Ratios of energies of higher excited states to that of the first excited state. In each isotope the ratio of the energy of some higher excited state (labeled with its spin) is compared to the energy of a first excited level with spin  $2+$ . A second level with spin  $2+$  is also present in these nuclei, and the ratio of its energy to that of the first excited level is labeled  $2+$ .

Points for isotopes of the same element are connected by straight lines labeled with the atomic number. The abrupt decrease of  $E_1$  between 88 and 90 neutrons contrasts with the gradual increase at  $110 < N < 116$  ( $Z=76$ ). Figure 7 presents a section of Figure 6 in the lower part and the ratios  $E_n/E_1$  in the upper part. Here  $E_n$  denotes the energy of the  $n$ th excited state with even spin and parity. The lines connecting the points are labeled with the spin and parity of the  $n$ th excited state. It is seen that the values  $E_n/E_1$  for the  $4+, 6+,$  and  $8+$  states decrease gradually from the rotational ( $\text{Hf}^{180}, \text{W}^{182}$ ) to the near-harmonic region (Pt), whereas for the second  $2+$  state  $E_n/E_1$  decreases rapidly and crosses the value for the  $4+$  state just beyond  $\text{Os}^{190}$ . The transition probabilities for the  $4+ \rightarrow 2+$  transitions in  $\text{Os}^{186}$  and  $\text{Os}^{190}$  were studied and compared with the ground state transitions  $2+ \rightarrow 0+$ . Systematic deviations of the values  $|M|^2_{4 \rightarrow 2}/|M|^2_{2 \rightarrow 0}$  from the value observed in  $\text{Hf}^{180}$  were found. The latter agrees well with the value 10/7 predicted by Bohr and Mottelson's theory for rotational nuclei.

The  $K$ -capturing Ir isotopes populating the Os levels were further investigated. As several of these isotopes have very similar half-lives, it was necessary to study the complex gamma spectra of all Ir isotopes from  $\text{Ir}^{186}$  to  $\text{Ir}^{190}$ . This work was done with the aid of a double-focusing spectrometer in collaboration with scientists at Harvard University. Knowledge of the beta spectra of the even Os isotopes, which were studied with the Brookhaven intermediate-image spectrometer, allowed some gaps to be filled in Way and Wood's graphs of total disintegration energies.

The question of nuclear shape has been the concern of some theoretical investigations from various aspects, such as the effect of the shape on the scattering of low energy neutrons by nuclei, or alternatively on the process of nuclear energy release known as internal conversion. In the latter case, however, the calculations show that the effects to be expected are exceedingly small.

The design and testing of a new detecting system for the intermediate-image pair spectrometer were completed. With the new detector the efficiency of collecting light from the scintillators has been improved by a factor of 10 over that of the old system, and this allows the coincidence circuit to be used at a resolving time of 1  $\mu\text{sec}$  with  $\approx 100\%$  coincidence efficiency. The instrument has been used at 1.25% resolution in conjunction

with the Van de Graaff to study the positron-electron pairs occurring in the beta decay of  $\text{N}^{16}$  (half-life 7.4 sec), the purpose being to search for beta decay to the 6.06-Mev  $0+$  first excited state of  $\text{O}^{16}$ . Most of the decays proceed to the 3-sec excited state at 6.14 Mev followed by a gamma-ray transition to the ground state. The 6.14-Mev internal pair line was found, but there was no evidence for a 6.06-Mev nuclear pair line component. It was possible to place a lower limit of 8.2 on the  $\log ft$  value of a beta-ray branch of  $\text{N}^{16}$  to the 6.06-Mev state of  $\text{O}^{16}$ .

The intermediate-image spectrometer has also been used for a number of studies of radioactive isotopes. The first known electric monopole cross-over transition from a  $0+$  second excited state was discovered (in  $\text{Ge}^{70}$ ) by means of a coincidence measurement. A spiral baffle system was constructed having a rejection ratio of  $\approx 5 \times 10^5$  against electrons of sign opposite to those being focused. It was used in the measurement of positron spectra occurring in the decays of certain isotopes of Ir mentioned above and in the decays of the ground and isomeric states of  $\text{Eu}^{152}$ .

The charged particles accelerated in the Van de Graaff accelerator have been used to study the details of a number of nuclear interactions. In view of the theoretical and experimental verifications of the finding that parity is not conserved in weak interactions, it becomes of interest to see whether the conservation of parity is still valid in strong interactions. In order to perform such a test, three types of experiments were carried out with charged particles from the Van de Graaff accelerator. One experiment consisted of a search for violation of the absolute selection rules in heavy particle transitions between states of known spin and parity. The case examined was the reaction  $\text{He}^4(d, \gamma)\text{Li}^6$  with the deuteron bombarding energy selected so as to induce resonant capture into the  $0+ 3.56$ -Mev level of  $\text{Li}^6$ . Such capture is completely forbidden by the usual spin and parity selection rules; thus the appearance of any 3.56-Mev gamma radiation would indicate a violation of the rules. No such radiation was observable. A second type of experiment dealt with the longitudinal polarization of heavy particles or the circular polarization of gamma radiation emitted from initially unpolarized systems in certain nuclear reactions. Polarization should not occur if parity is conserved. Two experiments in this class were carried out, one on the 2.14-Mev gamma-ray from

the first excited state of  $B^{11}$  formed in the  $B^{11}(p,p')B^{11*}$  reaction and the other on the 7.12-Mev gamma-ray from the fourth excited state of  $O^{16}$  formed in the  $F^{19}(p,\alpha)O^{16}$  reaction. The results again indicated that parity is conserved in these reactions. In the third type of experiment a search was made for odd powers of  $\cos\theta$  in the angular distribution of gamma radiation from certain nuclear reactions where only even powers of  $\cos\theta$  should occur if parity is conserved. The  $1/2$ -first excited state of  $Li^7$  at 0.477 Mev was excited by the  $Li^7(p,p')Li^{7*}$  reaction, and the angular distribution of the 0.477-Mev gamma-rays was studied. In all the experiments cited above, it was shown that parity is conserved in strong interactions.  $\mathcal{F}$  being the amplitude of the parity *nonconserving* part of the relevant wave functions, the most sensitive of the above experiments showed that  $\mathcal{F}^2 \lesssim 3 \times 10^{-8}$ .

Energy levels in  $C^{14}$  and  $N^{14}$  were examined by bombarding carbon targets enriched in  $C^{13}$  with deuterons and protons accelerated in the Van de Graaff accelerator. Various measurements were made on the gamma radiation emitted by means of scintillation detectors. The purpose was to see whether certain of the energy levels in these two members of an isobaric triplet could be identified as the lowest corresponding states of  $T=1$ . It was shown that the 6.09, 6.72, 6.89, and 7.35-Mev levels in  $C^{14}$  and the 8.06, 8.70, 8.90, and 9.51-Mev levels in  $N^{14}$  are very probably the  $T=1$  odd-parity states which correspond to each other, and that these states agree with shell-model calculations based on the known positions and properties of the four low-lying  $T=1$  states of  $N^{16}$ .

The proton beam from the Van de Graaff accelerator was used in an experiment designed to provide an accurate measurement of the polarization of protons elastically scattered from  $He^4$ . In this experiment the degree of polarization at three energies was measured by means of a specially constructed polarizer-analyzer scattering chamber. The right-left asymmetry in a second scattering of protons from  $He^4$  was observed by detecting the protons with photographic emulsions. The accuracy of these measurements is greater than for any previously reported values, and the results obtained agree with theoretical calculations based on single scattering phase shifts.

An experiment in progress at the Van de Graaff is the investigation of the elastic scattering of high energy gamma-rays from heavy elements by using

the 6 and 7-Mev gamma-rays from the  $F^{19}(p,\alpha\gamma)O^{16}$  reaction. In this reaction the gamma-rays are Doppler broadened by  $\approx 0.1$  Mev, and this band of radiation incident on a scattering material can excite, by resonance, any energy levels of the scattering nucleus which fall within the band. By measuring the cross section for resonant scattering in lead, the ratio of level width to level spacing,  $\Gamma/D$ , was found to be of the order of  $10^{-4}$ , in agreement with the value obtained by using bremsstrahlung.

Several nuclear interactions have been studied by guest scientists from Columbia University using the charged particles accelerated in the BNL 60-in. cyclotron. With protons in the energy range of 10 Mev, the angular distribution of the alpha-particles from the reaction  $Al^{27}(p,\alpha)Mg^{24}$  has been measured and compared with the Butler theory, with the conclusion that direct interaction processes play a substantial role in this reaction. On the other hand, the variation of the cross section with energy is unexpectedly rapid, which indicates that a resonance and hence compound nucleus processes are also involved. There is probably an interference between the two processes. The deuteron beam was used to study the angular distribution and cross section of the  $N^{15}(d,\alpha)C^{13}$  and  $N^{14}(d,\alpha)C^{12}$  reactions. In both cases the angular distributions are consistent with and have been fitted to a direct interaction theory.

The Columbia scientists have also studied the  $He^3(d,p)He^4$  reaction with the objective of finding an excited state in  $He^4$ . In considerable investigation, no such state was found. In particular, there was no confirmation of evidence for such a state at 22 Mev as reported by a group in the USSR. A new counter capable of better deuteron-proton resolution has been tested and will be used to re-examine the problem.

A  $\mu$ sec time-of-flight technique has been applied to the study of  $(p,n)$  reactions in  $K$ ,  $Ca^{48}$ ,  $Ti$ ,  $Cr$ ,  $Mn$ ,  $Fe$ ,  $Cu$ ,  $As$ , and  $Rb$  with the phase-focused 3-Mev proton beam from the BNL 18-in. cyclotron. The neutron spectra resulting from these reactions gave data from which the ground state  $Q$  values and some of the energy levels in the residual nuclei have been determined. The reactions in  $K$  and  $Fe$  yielded only one neutron group each, which corresponded to the ground states in  $Ca^{41}$  and  $Co^{57}$  respectively. In addition to the ground state  $Q$  values, the energies were determined for one excited state in each of the nuclei

$\text{Mn}^{53}$ ,  $\text{Mn}^{54}$ , and  $\text{Sr}^{85}$ , two excited states in  $\text{Zn}^{65}$ , three excited states in  $\text{Sr}^{87}$ , and four excited states in each of the nuclei  $\text{Sc}^{48}$ ,  $\text{V}^{49}$ ,  $\text{Fe}^{55}$ , and  $\text{Se}^{75}$ . The angular distributions of the neutron groups were observed to be essentially isotropic in the  $\text{Ca}^{48}(p,n)\text{Sc}^{48}$  and the  $\text{Cu}^{65}(p,n)\text{Zn}^{65}$  reactions.

### NEUTRON PHYSICS

Studies of the interactions between neutrons and atomic nuclei continue to yield information important to the better understanding of the properties of matter. In a continuing program of research in neutron physics at the BNL reactor, the slow chopper is used for experiments with very low energy neutrons. This chopper breaks up the neutron flux from the reactor into a succession of short pulses so that neutrons of different energies may be separated by time-of-flight technique. The "cold" neutron facility at the top of the reactor has been in normal operation during the past year, and the study of the modes of vibration of various crystalline materials has continued. Germanium has been extensively studied and is of special interest because of its semiconducting properties. The interaction of the low energy neutrons with the crystal is used to investigate both optical and acoustical modes of vibration. Figure 8 shows typical experimental data. Two strong peaks are evident in this figure, one of which corresponds to those neutrons which have gained energy from an optical mode of vibration in the crystal lattice, and the other to those neutrons which gain energy from an acoustical mode of vibration.

The "cold" neutron facility and the slow chopper are also being used for measurements on a series of metallic hydrides which might be useful as moderator materials in reactors. In these materials, hydrogen, which has a high neutron cross section, is tightly bound in a lattice of metal atoms. As neutrons are slowed down in such a system, the cross section exhibits sharp maxima at energies corresponding to the optical modes of vibration. The energy of a mode of this type in  $\text{ZrH}$  was reported last year. Other measurements are being made to investigate the optical vibrations for a series of metallic hydrides which have various ratios of hydrogen to metal atom concentration.

The research program with the fast chopper at the BNL reactor is a continuing study of total cross sections for neutrons in the energy range

from thermal to several kev. Many data have been obtained and analyzed for the purpose of testing different theoretical models of the nucleus, especially the optical model as modified for aspherical nuclei. "Area" methods of analysis have been applied to obtain the usual parameters of levels, such as neutron and radiation widths, but values of the spin have generally been assumed.

A program has been started to study the spectrum of the gamma-rays resulting from neutron capture which will yield information as to the spin of the levels in certain favorable cases. The method depends upon the ability to identify transitions between levels by making a pulse height analysis of the response of  $\text{NaI}(\text{Tl})$  scintillators to the gamma-rays emitted following neutron capture. The method has been applied to the neutron resonances in the reaction  $\text{W}^{183}(n,\gamma)\text{W}^{184}$ . The compound states formed by slow neutron capture have spins 1- or 0- and gamma-ray transitions to the ground state are strongly allowed for  $J=1$  and forbidden for  $J=0$ .

The time-of-flight technique was used to select the neutron energy corresponding to the resonances in  $\text{W}^{183}$ , and the gamma radiations were detected by four scintillators placed around the tungsten sample, but out of the direct neutron beam. The pulses from the detectors were analyzed with a single-channel pulse height analyzer and fed into the 1024-channel time analyzer normally used with the fast chopper. The scintil-

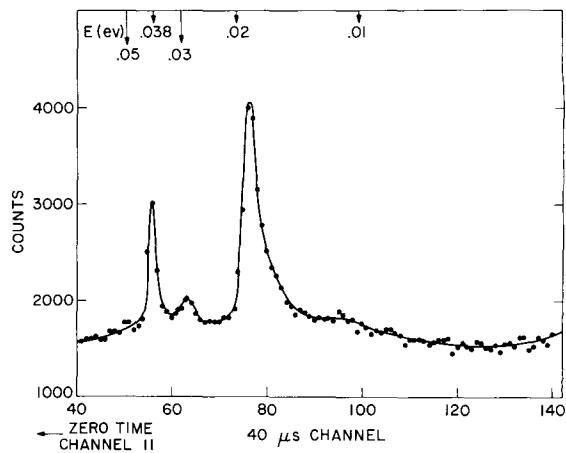


Figure 8. Spectrum of neutrons inelastically scattered by a single crystal of Ge; the peak at 0.038 ev results from gain of energy from an optical lattice vibration, whereas the peak at 0.019 ev results from the interaction with an acoustical vibration.

lators were shielded from the room by 6 in. of lead and from the sample by a mixture of boron carbide and paraffin 1 in. thick, so that neutrons scattered by the sample would not be captured by the iodine of the scintillators and thus result in a detector signal. With a narrow pulse height channel centered on 2.8 Mev, the known low energy tungsten resonances were observed by means of their capture gamma-rays as shown in the lower portion of Figure 9. The upper portion of this figure shows the effect of having the pulse height channel centered on 7.2 Mev so that only the highest energy transitions in  $W^{184}$  were detected. For the resonances in  $W^{183}$  only those with  $J=1$  should yield high energy gamma-rays; hence the 7.6 and 27.1-ev levels clearly have  $J=1$ . An analysis of the present results implies that only one of the unresolved resonances at 46.6 and 48.1 ev has spin 1. Further work is being done to extend the measurements to the other resonances of  $W^{183}$  as well as to other nuclides.

Additional measurements of the potential scattering cross section for nuclei have been obtained by the method of subtracting the contributions of

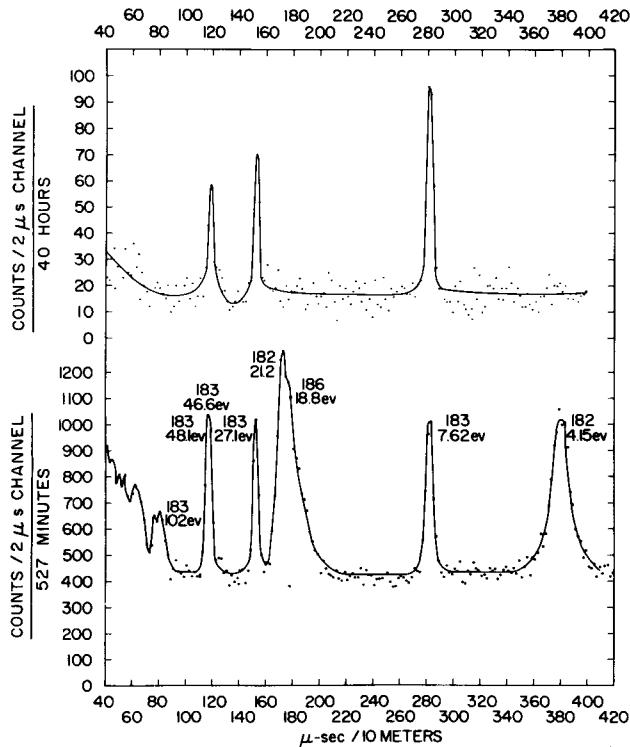


Figure 9. Time-of-flight spectrum of low bias and high bias channel settings.

the nearby resonances from the total cross sections at a given energy. The values of the nuclear radius found by using the relation  $\sigma_{\text{pot}} = 4\pi(R')^2$  are plotted against the mass number  $A$  in Figure 10. Also shown in the figure are curves giving the relation between these quantities as predicted by the "optical model" theory, which has been revised to take into account the aspherical form of

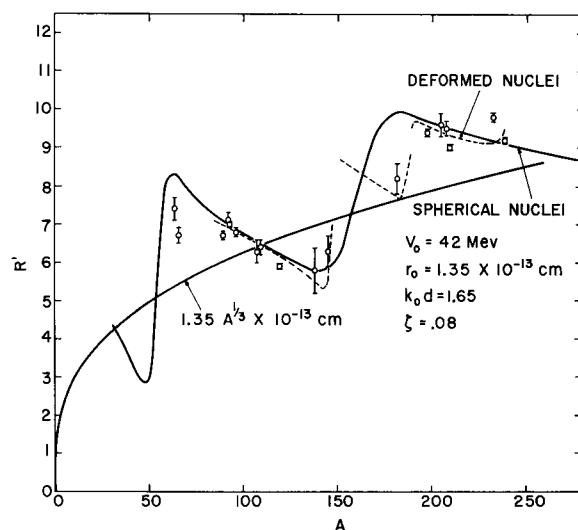


Figure 10. Potential scattering curve. The results of the measurements of potential scattering, displayed as  $R' = \sqrt{\sigma_{\text{pot}}/4\pi}$ , are shown together with the theoretical predictions based on the optical model of the nucleus. The spherical nuclei calculations are by Weisskopf, Porter, and Feshbach. The calculations for deformed nuclei were made by Wilets, Chase, and Edmonds, using experimentally determined deformation parameters.

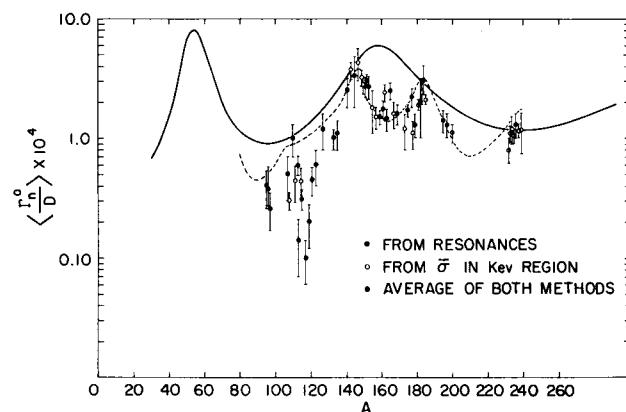


Figure 11. A comparison of the observed strength functions and the predictions of current theory based on the optical model. (See references under Figure 10.)

nuclei in the upper range of mass numbers. The agreement between computed and measured values gives strong support to this model of the nucleus and suggests that there is little need for further refinements of theory.

The program of measurements of the "strength function" for low energy neutron scattering has been continued. Theoretical investigation of this property for aspherical nuclei has given very good agreement with experimental points. The dashed curve in Figure 11 shows the theoretical results obtained by this modification of the "optical model" of the nucleus. The full line shows the values predicted by the theory of Feshbach, Porter, and Weisskopf, who considered all the nuclei as being spherical in form. The experimental points shown represent only the work done at Brookhaven by two methods; one involved the study of the parameters of individual resonances, and the other the measurements of an average cross section in the kilovolt region where the resolution does not separate the individual levels.

A study has been made of the distribution of level spacings for a number of nuclides, in particular those for which the spin is zero, since the difficulty of interpretation due to the presence of two spin states in the compound nucleus is thereby avoided. The study has brought out that there are fewer small spacings between levels than expected, an effect referred to as the "repulsion of levels." Guided by the distribution found for the zero-spin nuclei, the investigation was extended to include nonzero-spin nuclides, for which many

more data were available. Corrections had to be applied to the level distribution in order to take into account the possibility that resonances may be missed because of poor resolution, the small size of resonances, and the proximity of large resonances. Figure 12 shows the distribution of levels of the nonzero-spin nuclides plotted as a function of the ratio of the spacing between individual levels to the average spacing.

For some time a great need has been felt for a higher neutron flux to make it possible to obtain more precise cross section data, especially in the energy region from a few hundred ev to the kev range. To satisfy this demand, an arrangement has been made with Atomic Energy of Canada, Ltd., for Brookhaven to supply a fast chopper of the same design as the one in operation here to be set up at the heavy water reactor, NRU, at Chalk River, Canada. This reactor, which operates at 200 Mw, produces a neutron flux in excess of  $10^{14}$ , which exceeds that produced in the BNL reactor by several orders of magnitude. The auxiliary equipment, consisting of counters, 1024-channel analyzer, and most of the necessary manpower to operate the equipment, will be provided by Brookhaven. Some of the NRU personnel will actively participate with the BNL group in the work with this fast chopper.

The limited space available in the shield at the Chalk River reactor presents a difficult problem in redesigning all the accessories for the control and operation of the chopper. The schedule calls for completing preparation of the equipment by August and its installation and testing in the early fall of 1958.

The Compilation Group at Brookhaven has prepared a second edition of BNL 325, *Neutron Cross Sections*. The first edition of this report, which was itself the successor to the compilation AECU-2040, was prepared for the International Conference on the Peaceful Uses of Atomic Energy at Geneva in 1955. Since then it has been widely distributed by the Superintendent of Documents, Washington, D.C., and has become the standard reference handbook of neutron cross sections.

The second edition is now in press at the Government Printing Office, and 1000 copies are to be distributed at the Second International Conference at Geneva in September 1958. The need for the second edition is indicated by the fact that less than  $\frac{1}{3}$  of the material remains unchanged from the first edition.

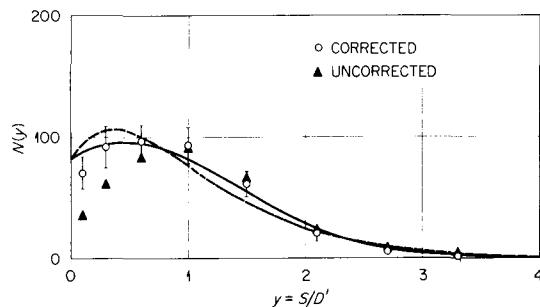


Figure 12. The level spacing distribution for the nonzero-spin nuclides plotted as a function of the ratio of the spacing between individual levels to the average spacing. The corrections were computed assuming a distribution of spacings given by the curves in the figure. The dashed curve gives the probability of obtaining any spacing regardless of the level spin. The solid curve shows the distribution as affected by Wigner's theory.

The second edition is divided into three sections, each with its own explanatory introduction and list of references. The first section contains 26 pages of tables of thermal cross sections, that is, reaction and scattering cross sections at a neutron energy of 0.0253 ev. This is the nominal energy of neutrons in a pile in equilibrium with a moderator at room temperature, and therefore knowledge of thermal cross sections is necessary in predicting the behavior of a reactor. Indeed, the fact that tables of thermal cross sections occupy this prominent place in the compilation is a reflection of the fact that the preparation of the compilation was originally motivated by the needs of reactor specialists.

The second section consists of 30 pages of resonance parameters. The energy levels (or resonances) in the compound nucleus which are investigated by neutron bombardment can be described by means of the Breit-Wigner formula with the appropriate resonance parameters. These parameters include the energy of the resonance, the various widths which make up the level (principally the neutron width, radiation width, and, where appropriate, the fission width), and the spin of the compound nucleus. A table of these parameters constitutes this section of the compilation.

The largest part of the compilation consists of 291 pages of curves of cross sections as a function of energy. Although total cross sections make up the bulk of the curves, there are also curves of the various partial cross sections - radiative capture, scattering, fission, and so on. The curves cover the energy region from  $<5 \times 10^{-4}$  ev up to 100 Mev, and include data on nuclei from hydrogen to Am<sup>243</sup>.

This new compilation contains all data, unpublished as well as published, received up to May 1958. The policy of listing only the "best values" for the various quantities has been continued from the first edition; that is, rather than listing all the measurements ever made of a particular cross section, the data are all carefully evaluated by the compilation group and the best value - not necessarily a simple weighted average - is chosen. The data in the second edition of BNL 325 were obtained from all the laboratories in the United States doing neutron cross section measurements, as well as from other laboratories in many countries throughout the world.

Although the preparation of BNL 325 was originally planned to meet the needs of reactor

specialists, it has proved to be extremely useful to nuclear physicists, radiochemists, and, in fact, all scientists interested in neutron reactions. It is expected that the second edition of BNL 325, like the first, will be widely used in laboratories throughout the world as the primary source book for data on neutron reactions.

Crystal spectrometers are used at the BNL reactor to produce highly monoenergetic beams of slow neutrons and to analyze in fine detail the energy distributions of slow neutrons in cross section measurements. Because of the high energy-resolution attainable with a crystal spectrometer, it is possible to study neutron resonances and neutron cross sections over narrow energy ranges. The study of slow neutron resonances in separated rare earth isotopes has continued this year with use of the high resolution crystal spectrometer. An important objective of this program is to measure the resonance parameters sufficiently well to be able to determine whether the values of the radiation widths  $\Gamma_\gamma$  for the different energy levels of an isotope are equal or fall into two or more groups. The rare earth isotopes are suitable for such measurements, since numerous resonances are observable within the energy range of the crystal spectrometer. Previous work with Eu<sup>151</sup>, Hf<sup>177</sup>, and In<sup>115</sup> indicated two distinct groupings of the radiation widths.

The analysis of the Sm<sup>149</sup> data was completed and reported. The four radiation widths measured were within 6% of the mean value. The low energy capture gamma-ray spectra were also studied with a scintillation spectrometer at the first two resonant neutron energies. No significant changes were found in the spectra for these two resonances, which is in agreement with the results of the Yale group.

Careful study of the Eu<sup>151</sup> and Eu<sup>153</sup> resonances has shown that for Eu<sup>151</sup> the previously reported grouping of  $\Gamma_\gamma$  into two sets is not correct. The four Eu<sup>151</sup> resonances at present analyzed give  $\Gamma_\gamma = 0.090 \pm 0.003$  ev, which differs markedly from the earlier result obtained with poorer resolution and natural Eu. Three more resonances are being analyzed for this isotope. In Eu<sup>153</sup> six resonances have been analyzed, five of them giving  $\Gamma_\gamma = 0.093 \pm 0.003$  ev; however, the sixth resonance at 6.16 ev gives  $\Gamma_\gamma = 0.130 \pm 0.003$  ev. Since this resonance is well isolated from the others, and since it is well fitted by the single-level Breit-Wigner formula, it is thought to be a single resonance. If this is true,

then there may well be two groups of radiation widths in  $\text{Eu}^{153}$ . Data have also been taken on Dy isotopes and are in the analysis stage. Studies are also being made of the low energy gamma-ray spectra associated with neutron capture for both  $\text{Eu}^{151}$  and  $\text{Eu}^{153}$ .

The low energy fission cross section data for  $\text{U}^{235}$ , obtained with the crystal spectrometer, have been analyzed in terms of the one-fission-channel multilevel formula of Reich and Moore. Results indicate that even though the single-channel fit is not perfect, it is sufficiently good to warrant the inference that at most a few fission channels are important. In this fit it is assumed that there is a large resonance at negative energy which is in the spin state opposite to that of the first two resonances. Vogt at Chalk River has used the BNL data and his multilevel analysis, which employs a few level many-fission-channel formula, to obtain a slightly better fit. The results indicate a smaller resonance at negative energy, and one which is in the same spin state as the first two positive energy resonances. This emphasizes the need for determination of the spins of the resonances to remove ambiguities in the analysis.

Measurements were made of the second-order contamination in the beam of Bragg-reflected neutrons used in the high resolution spectrometer. The calculated crystal reflectivities of Holm were verified. A simple method was worked out which enables the second-order component to be found by using the calculated reflectivities, and the open-beam count rate spectrum as a function of energy.

The program for measurement of the angular momenta associated with neutron resonances has continued as the major effort of the crystal spectrometer group. The components of a two-stage magnetic refrigerator, designed to operate at  $0.01^\circ\text{K}$  or lower for periods of several hours, have been built and tested. Two-stage adiabatic demagnetizations have been made, but the temperature achieved, although below  $0.1^\circ\text{K}$ , is uncertain. Very favorable operation of the first superconducting heat switch (Pb) was obtained, which indicated that, after the initial demagnetization, exchange gas is not necessary. Work is continuing to improve the design of the second stage with respect to cycle time, temperature measurement, low temperature reservoir capacity, and the provision of ripple-free magnet currents. A new cryostat is nearing completion which will minimize

vacuum and heat leak difficulties experienced with the first cryostat.

By using a magnetized Co crystal in a neutron spectrometer, highly polarized beams of neutrons have been obtained with adequate intensity for transmission measurements in the ev region. Tests are being made to determine whether a constant-deviation polarized-neutron spectrometer is feasible, since such a device would eliminate the complications inherent in a movable cryostat. It is expected that both spectrometer and cryostat design will be finished shortly, and that the combination will be installed at the reactor toward the middle of FY 1959.

Temperature measurements have been made with a thermometer which indicates the Johnson noise developed across a wire-wound resistor placed in the cryostat. With a comparatively crude circuit, the range  $300^\circ$  to  $0.9^\circ\text{K}$  was covered with excellent results. Modifications are being made which should allow substantially lower temperatures to be measured absolutely.

Neutron time-of-flight measurements have been made by exploiting a pulsed beam from the 60-in. cyclotron. For work up to a few hundred ev the ion source is pulsed to give a  $0.6\text{-}\mu\text{sec}$  burst with a peak current of 1 ma. Detection by the self-indication method has been used, i.e., neutrons are detected in the sample of interest by the  $n\text{-}\gamma$  process. The analysis of some results obtained by this method for neutron energies of a few hundred ev has been completed for Ag. Analysis of similar data for Au, W, Ta, and Mo is proceeding.

The use of the 60-in. cyclotron in connection with neutron time-of-flight measurements in the Mev region requires that a combination of ion source pulsing, the natural beam bunching, and deflector pulsing be used to produce single bursts of a few  $\mu\text{sec}$  duration at a peak current of 10 ma or greater. A method has been devised to obtain from the cyclotron single pulses of high energy particles widely separated in time in the  $\mu\text{sec}$  region. These pulses can then be used to produce short bursts of neutrons, and time-of-flight spectroscopy can be applied in the 2 to 25-Mev range of neutron energies. The method has been tested, and the neutron cross section values obtained compared favorably with previous measurements in this energy region. The broad energy spectrum of the neutrons, produced by the high energy particles from the 60-in. cyclotron, makes the measurements more complicated than those which can

be made with the monoenergetic neutron pulses available with lower energy sources. However, in certain types of neutron interactions it is advantageous to exploit the higher neutron energies in spite of the disadvantage introduced by the broad energy spectrum.

A  $\mu$ sec time-of-flight technique has been applied to the study of the scattering of 2.2-Mev neutrons from targets of Fe,  $Pb^{206}$ , and Y. The 3-Mev phase-focused proton beam from the BNL 18-in. cyclotron was used to produce high intensity neutron pulses from the  $T(p,n)He^3$  reaction. The angular distributions of the elastically scattered neutrons have been measured for the above elements. The angular distributions of the inelastically scattered neutrons from the 0.845-Mev level in  $Fe^{56}$  and from the 0.803-Mev level in  $Pb^{206}$  have been carefully studied and were found to be symmetric about  $90^\circ$  in the center-of-mass system. The angular distributions of the inelastically scattered neutrons from levels at 1.34 and 1.43 Mev in  $Pb^{206}$  and from levels at 0.913 and 1.53 Mev in Y have also been measured, and these also appear to be symmetric about  $90^\circ$  in center-of-mass coordinates. The inelastic scattering from  $Pb^{206}$  shows evidence of a new level at 1.15 Mev.

### ATOMIC AND MOLECULAR PHYSICS

The techniques of atomic beams and paramagnetic (electron) resonances are being used in the continuing nuclear moments program. The objectives of this program are to obtain highly precise measurements of the spin, magnetic, and higher moments of atomic nuclei. Particular emphasis is being placed on studies with radioactive nuclei.

The atomic beam apparatus has been modified to measure nuclear magnetic moments as well as nuclear spin. With this apparatus, studies will be made on the hyperfine structure anomalies in several of the radioactive alkali nuclei. The precision required for the magnetic moment measurement is at least one part in  $10^3$ , which implies an accuracy in determination of the atomic beam resonance of one part in  $10^7$ .

The instrument has been tested by making a measurement on stable  $Rb^{85}$ . The next objective is to make measurements on the 3-hr isomer  $Cs^{134m}$ . An attempt has also been made to determine the nuclear spin of  $Eu^{152m}$  (9-hr half-life), but it is necessary to develop further an oven capable of withstanding a temperature of  $1500^\circ C$  in order to carry out this experiment.

A commercial X-band spectrometer, recently received, has been used to study several specimens of irradiated polyethylene of interest to the Nuclear Engineering Department. Some work is in progress to confirm preliminary results obtained on the measurement of the nuclear spin of  $V^{49}$ . The spectrometer has also been used to examine irradiated seed for the presence of free radicals, this being a problem of interest to the Biology Department.

A program to investigate the magnetic moment of  $Na^{21}$  has been started at the BNL 60-in. cyclotron by a guest scientist from Princeton University. Because of the short half-life (23 sec) the experiment must be done within a few feet of the cyclotron target. The sodium is produced from  $Ne^{20}(d,n)Na^{21}$  and is then pumped (or blown) through a tube to the far side of a shielding wall, where it is lined up with an optical pump. Preliminary measurements have shown that the amount of sodium produced, the operating pressures, the transport, and background problems are such that the experiment is feasible.

### SOLID STATE PHYSICS

#### Structure of Solids

The neutron diffraction program of the Physics Department continues to be concerned mainly with structural studies on magnetic and ferroelectric crystals. Some studies are also in progress in which crystal structure is of secondary importance. These studies are directed at some fundamental questions in the theory of magnetism, the theory of the liquid state, and the character of crystal transitions.

The final refinement of the ferroelectric Rochelle salt structure has been completed, and the structure suggested last year confirmed. Among the many atomic displacements found in the structure (relative to the paraelectric symmetry), the most significant one appears to be that of one of the hydroxyl groups on the tartrate molecule. The experimental work has been followed by a theoretical local field treatment based on the proper structure. In the theory it was also possible to generalize Mueller's phenomenological theory of Rochelle salt, and to describe the dielectric properties of the clamped crystal.

A study is now in progress on the atomic Debye-Waller parameters for  $BaTiO_3$  in the cubic paraelectric phase. It is hoped that this work will re-

sult in a determination of the shape of the potential well for Ti. The thermal vibrations of Ba and Ti must be isotropic, but anisotropy may be present in the case of oxygen. According to a theory of Devonshire, the complete ferroelectric behavior of  $\text{BaTiO}_3$  can be worked out from a thorough knowledge of the nonferroelectric cubic phase.

Two additional ferroelectric structure analyses were started in conjunction with x-ray diffraction studies at Pennsylvania State University, on  $(\text{CH}_2\text{NH}_2\text{COOH})_3 \cdot \text{H}_2\text{SO}_4$  (triglycine sulfate) and  $(\text{ND}_4)_2\text{BeF}_4$ . The x-ray work on both these crystals is well along, but the neutron analysis is still in the stage of data collection.

Work on the  $\alpha\text{-Pb}(\text{N}_3)_2$  structure has temporarily been suspended. It was found that a three-dimensional attack on the problem would be necessary. A modification is now in progress of one of the spectrometers that will be convenient to use in the collection of nonequatorial data. The modified instrument will permit the use of equi-inclination techniques. Another instrumental improvement just completed is the modification of one of the single-crystal spectrometers for automatic programming. A sequence of observations can be programmed by setting appropriate instructions in a switch panel. This should add considerably to the efficiency of data collection.

In the thermalization of neutrons by hydrogenous materials, the hydrogen can be treated essentially as free for neutron energies  $> 0.5$  ev. For energies lower than this, the chemical or crystal binding of the hydrogen markedly influences the energy exchange. An experimental investigation has been made, in a cooperative program with General Atomic, of this low energy region for water, ice, Lucite, polyethylene, zirconium hydride, and magnesium hydride by using several techniques. The total cross section has been measured as a function of energy. This shows resonances in the metal hydrides due to the oscillations of the bound hydrogen and gives the energy of these bound levels. A method similar to that used in neutron diffraction has been used to study the average logarithmic energy decrement for the above substances. The metal hydrides give remarkable fluctuations in the variation of the decrement as a function of neutron energy, and these fluctuations can be related to the hydrogen oscillator levels. A third technique involving the time-of-flight of the neutron scattered at a  $90^\circ$  angle from various substances has been employed to study the scattering kernel in detail. Particular at-

tention has been given to water in an effort to study the ideas advanced independently by Nelkin of General Atomic and Brockhouse of Chalk River. This latter technique has also been used to study the scattering of neutrons by lead just below and just above the melting point and at a temperature of  $300^\circ\text{C}$  above the melting point. This study was made to check the new theory of neutron scattering by Vineyard. One of the simple models suggested by Vineyard, the diffusional one, does not describe the temperature dependence of the neutron scattering for lead, whereas the gas model does come near to agreeing with the experiments.

The study of short-range order in the brasses is of fundamental interest but is complicated by the similarity of the scattering powers of copper and zinc for both x-rays and neutrons. The neutron scattering lengths of  $\text{Cu}^{63}$  and  $\text{Cu}^{65}$  have been measured and are significantly different from those of natural copper. The  $\text{Cu}^{65}$  isotope has the larger scattering length, and the short-range order scattering in brass with  $\text{Cu}^{65}$  as the copper constituent is  $6\frac{1}{2}$  times as intense as that with natural copper. The absorption and incoherent cross sections for natural copper and its isotopes have been determined. With the above enhancement of short-range order scattering, the order-disorder transformation in  $\beta$ -brass becomes experimentally accessible to neutron diffraction techniques. The diffuse scattering of monochromatic neutrons from a single crystal of  $\beta$ -brass isotopically enriched in  $\text{Cu}^{65}$  has been measured at three temperatures above the critical temperature of  $468^\circ\text{C}$ . Analysis of these data is in progress. At each temperature Fourier inversion of the scattering gives the local correlation of atoms, i.e., the degree to which an atom influences the distribution of atoms on neighboring sites. These data will serve as criteria for establishing the validity of the various theories of such cooperative phenomena.

The effect of temperature on diffuse short-range order scattering has been neglected in the past. In connection with the above experimental work, a concise expression for the intensity of the diffuse scattering from binary alloys has been developed in which the effects of temperature are explicitly included. Basically this expression is the same as that derived previously by neglecting temperature effects, except that the short-range order parameters are modified by a Debye factor which depends on the correlation distance. This temperature effect cannot be neglected in measurements of short-range order at elevated temperatures and at high

scattering angles. For example, failure to include this effect places the published value of the first-neighbor short-range order coefficient in  $\text{Cu}_3\text{Au}$  at 405°C in error by  $\approx 25\%$ , and the error is larger for the more distant neighbors and the higher temperatures.

Concurrently with the study of short-range order in  $\beta$ -brass, an attempt was made to detect short-range order in a single crystal of  $\alpha$ -brass annealed three months at 125°C. The lattice parameter and hydrostatic density changes which have been attributed to changes in the degree of local order were substantiated. However, the neutron diffuse scattering did not show conclusive evidence of the existence of local order. Greater sensitivity is possible if the crystal is made from copper isotopically enriched in  $\text{Cu}^{65}$ . A program based upon this approach is being planned for a further investigation of this problem.

Investigation of possible order in several iron alloys was made by neutron powder diffractometry. The equiatomic FeCr alloy showed no signs of long-range order after 100 hr annealing at 475°C, in contradiction to some x-ray results reported in the literature. The equiatomic FeCo alloy showed the usual ordered CsCl structure after long annealing at low temperature. The compositions  $\text{Fe}_3\text{Co}$  showed superlattice reflections that indicate either a homogeneous poorly ordered FeCo structure or a mixture of a small amount of ordered FeCo with a disordered iron-rich phase.

The study of magnetic form factors with the polarized beam spectrometer has been continued. Some interesting conclusions have been drawn from comparison of the neutron results on iron, nickel, and cobalt and the similar measurements with x-rays made by Weiss and DeMarco. For iron, the x-ray and neutron results are in agreement, while those for cobalt and nickel show differences both in the absolute magnitude and in the shape of the form factors as a function of angle. Furthermore, since the x-ray results (after subtraction of the argon core) represent the contributions of all the 3d localized electrons, while the neutrons scatter only from the unpaired electrons, the two sets of measurements allow separate calculations of the radial distribution for the two spin states. This has been done for nickel, and shows a separation of several hundredths of an angstrom between the peaks of the electron densities for the two spin states.

A further study has been made on the neutron scattering of  $\text{Fe}_3\text{Al}$  at low temperatures. This ma-

terial was previously shown to possess iron atoms with two distinct magnetic moment values. The low temperature neutron measurements definitely rule out the possibility that this behavior could be explained by an antiferromagnetic coupling between next-nearest-neighbor iron atoms via an aluminum atom. Additional work on  $\text{Fe}_3\text{Al}$  single crystals with polarized beams also demonstrated that the form factors of the magnetic electrons surrounding the two different iron sites are essentially the same. All this evidence tends to support the view that there is a transfer of electrons between the iron atoms and the nearest-neighbor aluminum atoms.

As part of a continuing program on the spin structures of magnetic oxides, the mixed systems  $\text{MnFe}_2\text{O}_4$ - $\text{MnCr}_2\text{O}_4$  and  $\text{FeTiO}_3$ - $\text{Fe}_2\text{O}_3$  have been investigated. The former is of interest because of the wide differences between the observed magnetic moment values and those predicted by the Néel model. Measurement of the individual sublattice magnetizations showed that 1) it is the octahedrally coordinated sites which always possess a magnetic moment less than expected; 2) the temperature dependences of the sublattice magnetizations are in agreement with those calculated by J.S. Smart from the Néel theory; 3) all the magnetic scattering data can be fitted to an anti-parallel arrangement for the two sublattices, which excludes ordered triangular spin configurations; and 4) there is a definite connection between the difficulty in saturating the material and the anomalous behavior of the octahedral sublattice magnetization.

The  $\text{FeTiO}_3$ - $\text{Fe}_2\text{O}_3$  system is of interest because of the appearance of ferromagnetism in a system combining two antiferromagnetic substances. It was shown that  $\text{FeTiO}_3$  possesses a magnetic unit cell twice as large as the chemical cell. Such a large cell calls for the existence of an antiferromagnetic coupling between the  $\text{Fe}^{2+}$  atoms over a considerably larger distance than had been expected. For the mixed structures containing more than 20%  $\text{Fe}_2\text{O}_3$ , the presence of ferromagnetism has been shown to come from an ordered antiparallel arrangement of layers of  $\text{Fe}^{2+}$  atoms and mixed  $\text{Fe}^{2+}\text{Ti}^{4+}$  layers. When the material is quenched, this order is partially or fully destroyed, which leads to a lowering of the total moment.

#### Radiation Effects

The other major activity in solid state physics is the study of defects in crystals. Radiation effects

and other departures from perfect periodicity are under investigation with many diverse techniques.

Very little work has been done on the dynamics of the production of crystalline defects by radiation at temperatures at which the defects tend to disappear by various annealing mechanisms such as direct recombination of vacancies and interstitials, migration to dislocations, etc. Simultaneous production and annealing processes can now be studied by the technique of radiation-enhanced diffusion. It was pointed out in last year's report that radiation-enhanced diffusion is observable in the  $\alpha$ -brass system since it leads to increased short-range order. The phenomenon of enhanced diffusion in this system has now been studied in a quantitative way. A simple theory of radiation-enhanced diffusion has been worked out which describes the dependence of this enhancement on flux and temperature under steady-state conditions. The theoretical study also shows that the measurement of enhanced diffusion as a function of temperature can indicate the mechanism by which defects are removed from the lattice. The enhanced diffusion rate during irradiation in the

Brookhaven reactor has been measured in  $\alpha$ -brass at several temperatures in the range from 0° to 190°C. This enhancement is independent of temperature in the range from 0° to 150°C, in excellent agreement with the theoretical predictions for the case when the radiation-induced defects finally disappear at internal surfaces. The data from 150° to 190°C also follow very accurately the predictions of this simple theory. This new technique will be applied to a number of problems.

A related study is concerned with the effect of radiation on the martensite transformation in an iron-nickel (25% Ni) alloy. A high temperature irradiation produces resistivity changes which are indicative of phase separation, probably by means of enhanced diffusion. Irradiation at lower temperatures inhibits the subsequent martensite transformation as well as producing some transformation during irradiation. The data show that a neutron exposure of  $4 \times 10^{17}$  nvt epicadmium lowers the martensite start temperature,  $M_s$ , in this alloy by  $\approx 6^\circ\text{C}$ . The decrease in  $M_s$  is believed to be due to damage of the same type as that which raises the critical shear stress, since it is known

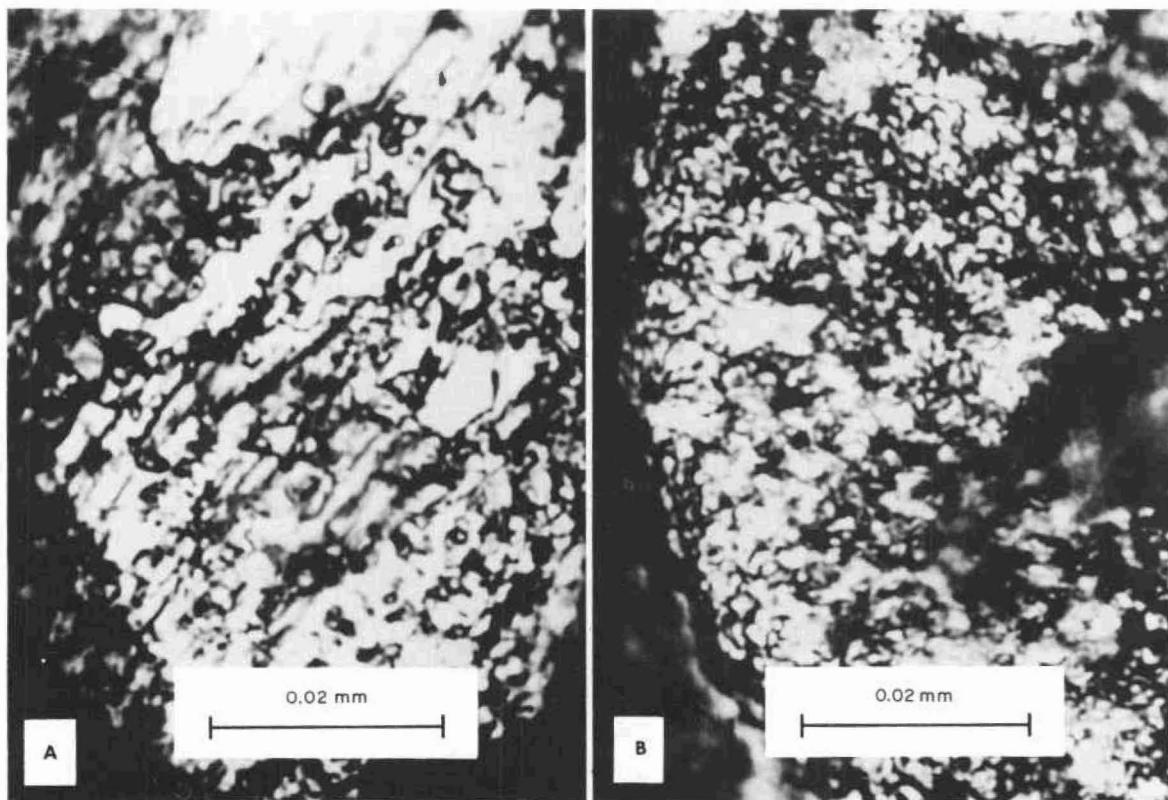


Figure 13. Diamond surfaces after grinding, *A*, unirradiated; *B*, irradiated.

that the  $M_s$  temperature is structure sensitive. A low exposure experiment indicated that the effect is not due to a change in elastic modulus, since such changes should saturate below  $10^{13}$  nvt and since there was no change in  $M_s$  for such an exposure.

During irradiation, changes occur which have been studied by means of resistivity and magnetic measurements. Analyses of these results indicate that over 1% of the alloy transforms during irradiation. Moreover, in specimens that have been partially transformed prior to irradiation, some of the strain induced by the transformation is recovered during subsequent irradiation. This recovery occurs in spite of the fact that radiation damage is occurring concurrently. This is believed to be the first clear-cut observation of irradiation-induced strain recovery. The mechanism of the recovery is not yet clear.

Heavily irradiated diamond has been investigated further by both x-ray techniques and mechanical measurements. The heavily irradiated structure is highly strained, which causes the interference effects to be weakened and produces an x-ray pattern of an amorphous-like structure. Diffraction data are complete, and the integral Fourier inversion for the radial distribution of the number of atoms between  $r$  and  $r + dr$  is being computed for three diamond samples with lesser damage. No sudden onset of the amorphous-like structure is evident, but a gradual degradation of the interference effects with increasing irradiation is observed. For small irradiations the x-ray reflections are shifted, which indicates a lattice expansion in agreement with density measurements, and the intensities are attenuated with increasing scattering angle. For moderate irradiations the peak shifts are more pronounced, a diffuse intensity peaking near the undamaged line position builds up, and the reflections at large scattering angles are replaced by broad diffuse peaks. The hydrostatic density is characteristically less than that determined from the lattice expansions. For heavy irradiation the entire pattern is diffuse.

The mechanical properties of irradiated diamond have been investigated by measuring the amount of wear of neutron-irradiated diamond against silicon carbide abrasive and comparing it with that for unirradiated diamonds. The volume removed from diamond phonograph needles was calculated geometrically from photomicrographs of four groups of six diamonds irradiated with different total fluxes, and an average was taken for

each group. For an irradiation of  $2.6 \times 10^{18}$  nvt (fast) the hardness was 50% of the unirradiated hardness, and for  $7.8 \times 10^{20}$  nvt (fast) the hardness was 20%. The densities were measured by flotation in warm thallium malonate solution to an accuracy of about  $\pm 0.7\%$ . For the  $2.6 \times 10^{18}$  irradiation the density had decreased by 1%, and for the  $7.8 \times 10^{20}$  irradiation by 4%. The decrease of cohesive energy of an irradiated diamond was estimated crudely from the number of broken bonds and the strain energy of the density change. The calculated fractional decrease in cohesive energy is about one-half of the fractional decrease in hardness. Thus, diamond becomes softer on irradiation, contrary to the response of most metals. In the case of metals, dislocation motion is held up by the radiation-induced defects. Thus, the hardness of diamond is apparently controlled by the cohesive energy, which decreases upon irradiation as indicated above, rather than by dislocation motion. Further evidence for this picture is provided by photomicrographs of the ground surfaces. The picture of the unirradiated surface (Figure 13) reveals scratches and chips removed around the scratches. Regions where there are no scratches exhibit a very smooth surface. This is what is expected if the polishing action is by attrition only, that is, the fine chips are removed more readily at existing faults. In the case of the irradiated diamond, also shown in Figure 13, a substantially different material is exposed to the abrasive, since the density of faults is higher and the cohesive energy is decreased.

An investigation has been carried out of the effect of reactor irradiation on single crystals of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>. This was primarily a continuation of the use of long-wavelength neutron transmission for determining the concentration and types of defects produced in solids by high energy particles. The aluminum oxide exhibited crystallographic stability to fast neutron irradiation at temperatures  $< 40^\circ\text{C}$ , and the results indicate a total number of defects  $\approx 40$  times less than that predicted by current theories. Correlation of the transmission with the macroscopic density changes was good. The fact that the fractional change in density was always greater than the fraction of defects present suggests that the lattice relaxation is outward around a vacancy as well as around an interstitial. Annealing of the material produced no decrease in the concentration of defects from room temperature to  $400^\circ\text{C}$ , a steady decrease from  $400^\circ$  to  $1250^\circ\text{C}$ , and irregular changes beyond  $1250^\circ\text{C}$ .

Annealing at 1800°C did not remove the coloring, although the density returned to its preirradiation value.

With the completion of the study of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>, attention has been turned toward a similar investigation of beryllium. Plans have been made to correlate changes in the resistivity with changes in the neutron transmission of the identical beryllium samples as a function of irradiation dose. This experiment, as well as many others, requires low temperature irradiation. The new low temperature irradiation facility is nearing completion. The in-pile sections have been completed and leak-tested. Because of the sensitivity of the Brookhaven reactor to the amount of in-pile foreign material, it has been found necessary to devise a method of filling the irradiation chamber with liquid nitrogen different from that previously employed. A devise for detecting the liquid level is now being perfected.

The neutron transmission experiments on Al<sub>2</sub>O<sub>3</sub> are closely related to the optical studies in progress on this material. One of the important color centers in  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> absorbs at 2040 Å. It has been shown previously that this band is formed by reactor irradiation but not by purely ionizing radiation. By careful annealing studies it has now been shown that upon heating the crystal the 2040-Å band is removed in two stages. The first or low temperature stage appears to be finished when the crystal is heated to  $\approx 300^\circ\text{C}$ . The second or high temperature component is not completely removed until temperatures in excess of  $1000^\circ\text{C}$  are reached. It is interesting to correlate this result with the measurements of radiation damage in Al<sub>2</sub>O<sub>3</sub> made by the long-wavelength neutron transmission technique. These neutron transmission measurements indicate that radiation damage does not begin to anneal out until the sample is heated to  $340^\circ\text{C}$ , which is close to the temperature at which the second annealing stage begins. The tentative conclusion is that the high temperature component represents removal of defects while the low temperature component is related to an undetermined electronic process (rearrangement of electrons). The activation energies associated with the first component are  $\approx 1$  ev, while 1.5 to 2.5 are obtained for the second group.

In another series of experiments on  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> samples were heated near their melting point and cooled in a controlled manner. Simply heating

$\alpha$ -Al<sub>2</sub>O<sub>3</sub> for several hours at 1800°C removes all the absorption present in the samples when received from the manufacturer, in the range 2000 to 10,000 Å. Since this treatment can increase the transmission at 2000 Å from  $\approx 50\%$  to that given by the Fresnel reflection loss, it is a useful and practical way of improving the ultraviolet transmission of Al<sub>2</sub>O<sub>3</sub> windows. Subsequent exposure of the heat-treated samples to ionizing radiation restores the coloring to the original level. Another result of these high temperature experiments is that the defect responsible for the 2040-Å band may be present, to a small extent, in unirradiated samples. These high temperature experiments will continue with particular emphasis on the possibility of "quenching in" defects. Annealing measurements on the radiation-induced color centers in very pure fused silica gave similar results. However, the fused silica data are extremely laborious to analyze, since the bands overlap considerably, and it may be some time before a reasonable understanding is obtained.

The cooperative program with R. Truell's group at Brown University utilizing ultrasonic techniques to study radiation-induced changes on mechanical properties has continued. The principal new results were obtained on glass samples that contain a few percent B<sup>10</sup>. When they are exposed to reactor irradiations, the radiation damage results from the B<sup>10</sup>(n,α)Li<sup>7</sup> reaction. Since this reaction proceeds with slow neutrons, the contribution to the damage from the usual fast neutron recoils is negligible during the 5 to 60-min exposures used. The damage from the above reaction is so severe that an irradiation of  $>60$  min often breaks up the sample into a dozen or so fragments. In the unirradiated glass the ultrasonic velocity increases with frequency, while, in contrast, in the irradiated material the velocity decreases with frequency. The way the velocity changes as a function of dose is irregular and not understood at the moment.

The strain patterns in boron-containing glass can be determined by photoelastic techniques. With the cooperation of C. Mylonas of Brown University the strain patterns in boron glass samples have been thoroughly analyzed. The main result is that the distribution of strain is in one to one correspondence with the distribution of the B<sup>10</sup>(n,α)Li<sup>7</sup> reaction, i.e., the absorption of slow neutrons. While this result is not unexpected, it does constitute a demonstration that, at least in

this material, the radiation damage is directly proportional to the number of damaging events.

The cooperative program with Picatinny Arsenal continues into its fourth year. This effort is primarily concerned with obtaining a fundamental understanding of the effects of radiation on explosives and related materials. One aspect of this program, the study of the thermal decomposition kinetics of lead styphnate monohydrate (an explosive used in detonators) which has been subjected to gamma-ray and reactor irradiations, has been concluded. Cobalt-60 gamma-ray irradiations up to  $1.8 \times 10^8$  r did not significantly alter the thermal decomposition curves. However, material subjected to reactor irradiations decomposed as much as three times as fast as unirradiated material, and the increase in rate was roughly proportional to the irradiation. The activation energy for the decomposition reaction was not changed by the irradiations. The conclusion is that the irradiation increased the number of decomposition nuclei but did not alter the decomposition mechanism. The apparatus used for this decomposition work has recently been greatly improved, and a series of experiments has been started to correlate the thermal decomposition of  $\text{KN}_3$  with the color center, x-ray, and microscopic measurements on this material.

Studies of the color centers formed in the azides by radiation continue, although this work is seriously hampered by our inability to grow really good crystals routinely. Several new crystal-growing techniques show considerable promise but need to be developed further. A large amount of data on the coloring of  $\text{KN}_3$  by gamma-rays at room temperature has been analyzed. Most of the observed absorption bands are probably color centers, although one broad band might be caused by colloidal potassium which aggregates in the crystal during irradiation. An experimental and theoretical study of this possibility is now under way. Studies of the gamma-ray induced coloring in  $\text{KN}_3$  are also being made with both irradiations and measurements carried out at liquid nitrogen temperature.

The x-ray study of radiation effects in sodium azide has been completed. Stacking faults are produced by mechanical deformation,  $\gamma$ -irradiations, and reactor irradiation. The azide molecule is decomposed (three molecular decompositions per 100 ev of energy absorbed) by ionizing radiation. The local stresses produced by decomposition pro-

duce dislocations and associated stacking faults similar to those produced by mechanical deformation. Annealing studies show recovery of the damaged state with return to stoichiometry of the irradiated crystal. Calculations of the energy absorbed in this material through ionization from x-rays and gamma-rays of the  $\text{Co}^{60}$  source, and from the various ionizing reactions during reactor irradiation, are in substantial agreement with the observed effectiveness of these radiative sources in producing damage in sodium azide. During the course of this study, the importance of the gamma flux induced in a sample in the presence of thermal neutrons by  $(n, \gamma)$  reactions was considered. The effect is important when the  $(n, \gamma)$  cross section is of the order of several barns, and is the primary mechanism of energy absorption in some cases. For example, the self-induced gamma flux in one  $\text{cm}^3$  of rock salt is  $3\frac{1}{2}$  times as intense as the pile gamma flux in a typical thermal reactor.

The increase in the oxidation rate of graphite by nuclear radiation has been discussed in previous reports. However, two variables, impurity content and heat treatment of the graphite, had not been studied in detail. Recently, experiments have been completed on unirradiated and irradiated graphite annealed at  $600^\circ$  and  $2800^\circ\text{C}$  and subsequently oxidized at  $300^\circ\text{C}$ . Annealing did not affect the oxidation rate of unirradiated graphite, which indicated that the impurities in unirradiated graphite do not cause catalytic burning: the impurity content was found to decrease during annealing at  $2800^\circ\text{C}$  from about 0.1% to about 0.001% without altering the combustion rate. Irradiated samples annealed at  $600^\circ\text{C}$  burned at about the same rate as unannealed irradiated graphite, but after annealing at  $2800^\circ\text{C}$  they burned at the preirradiation rate. Thus, the radiation effect was removed by annealing between  $600^\circ\text{C}$  and  $2800^\circ\text{C}$ .

### Theory

The major theoretical problems under active investigation in solid state physics, often closely related to the experimental research, are described in this section.

The effect of the regularity of atomic arrangement on the production of displaced atoms by irradiation of crystals has been considered. Because of a tendency for energy to be "focused" down closely packed lines of atoms, it is predicted that displacements may be somewhat harder to make

than had been contemplated in conventional theories, and the consequences of this have been worked out semiquantitatively. A calculation of the details of the displacement process with the aid of a large electronic computer is also under way.

It was suggested some time ago by Varley at Harwell that neutral interstitial halogen atoms may be produced by ionizing radiation in the alkali halides. There is some support for this idea based on optical and magnetic resonance experiments. A theoretical investigation of the properties of interstitial halogen has been started based on the classical treatment of an ionic crystal. In particular, the most stable configuration of an interstitial and the energy barrier for its diffusion are being determined. Preliminary results indicate that the chlorine interstitial in KCl resides in one of the cube faces and that the neighboring chlorine ions have suffered a large displacement. This configuration, obtained by minimizing the energy of the system (Coulombic, polarization, and repulsive terms), is in reasonable agreement with the available experiments.

A new approach to the scattering of slow neutrons by liquids has been found. By means of this approach the cross section for scattering with change of momentum and energy has been successfully factored into a part dependent on simultaneous correlation of atomic positions and a part dependent on the details of motion of a single atom in the liquid. Approximate theories of the latter motion give useful expressions for the cross section, and, conversely, measurements of neutron scattering by a liquid can now be used to determine the motion of an atom. The extension of these methods to molecular liquids is now being considered.

A study of the phases of Bloch functions has been continued. A relation between these phases and the crystal coordinate representation has been found. Analogous currents along lines of degenerate points in  $\mathbf{k}$  space cause small scattering effects. These are presumably responsible for the anomalous Hall effect in ferromagnetics.

The relative structure factor entering into the cross section for scattering of neutrons by spin waves in a normal spinel with zero  $A$ - $A$  and  $B$ - $B$

interactions has been found to be singular at the zero of the net spin for infinite magnon wavelength, in disagreement with the previously published result. The ratio of the new formula to the one used previously is about 3.5 for magnetite (for long magnon wavelengths). In connection with a proposed experiment (at Chalk River), the details of the scattering for the optical spin wave modes, including  $A$ - $A$  and  $B$ - $B$  interactions, are under investigation.

A study of the high temperature properties of the spin correlation function  $\gamma$  on the basis of the Heisenberg theory of magnetism has recently been initiated. The method used leads to rigorous expressions for the coefficients in a series representation of  $\gamma$  in powers of  $J/kT$  ( $J$  is the angular momentum). One of the objectives of the investigation is a comparison with the recent Elliott-Marshall theory, which is based on a molecular field approach (an extension of the constant coupling approximation), and which gave a general qualitative agreement with critical scattering experiments of Lowde. Quantitatively, however, there was considerable discrepancy between theory and experiment for  $T > T_c$ , where  $T_c$  is the Curie temperature (there were essentially no experimental data for  $T < T_c$ ). To date,  $\gamma$  has been obtained to order  $T^{-2}$  for the linear chain and the body-centered cubic lattice. The results indicate that further refinements of the molecular field approximation are not likely to resolve the discrepancy.

Another aspect of a high temperature theory of  $\gamma$  which appears to be promising is the determination of the Curie temperature by a method analogous to the Kramers-Opechowski method, which is known to fail for antiferromagnets. The failure is due to the fact that the susceptibility, which is infinite at  $T_c$  for ferromagnets, is bounded in the antiferromagnetic case. However, a suitably defined correlation distance should diverge at  $T_c$  independent of the sign of  $J$ . The approximation giving  $\gamma$  to  $O(T^{-2})$  leads to values of  $T_c$  very nearly that of the Weiss molecular field. For antiferromagnets  $T_c$  is found to be slightly larger than the ferromagnetic  $T_c$  in this approximation, the difference going to zero as the spin approaches infinity, all qualitatively in agreement with the result of the Bethe-Peierls-Weiss theory.

## Accelerator Development

The major concern of the Accelerator Development Department continues to be the design and construction of the 25-Bev Alternating Gradient Synchrotron. During the past year all structures which will house the machine complex were completed and subsequently occupied by the Department.

A contract was awarded for the fabrication of the main magnet coils and cores. Three cores and 16 pilot coils were shipped to Brookhaven for evaluation. After these units had been tested and approved, production releases for the balance of their respective contracts were given to the vendors. By the end of June, 47 cores and 242 coils had been delivered to Brookhaven.

The development of a high powered klystron for the linear accelerator rf source did not proceed as rapidly as had been predicted. Because of the tight linear accelerator schedule, it was decided to drop the klystron and substitute the immediately available French-Thomson Houston TH470 triodes. Fabrication of the eleven 10-ft-long sections of the linac tank is under way, and the first section has arrived on site. The assembly and contouring of the linac drift tubes are proceeding, but the final brazing operation must await completion of the hydrogen furnace facility. The initial testing of the 750-kv Cockcroft-Walton preaccelerator was completed, and the unit has been moved to its final location in the linac building.

The design of the radio-frequency accelerating system is progressing satisfactorily, and specifications for the 12 power amplifiers have been prepared to accompany the solicitation of proposals for fabrication. To date, 860 ferrite rings for the accelerating cavities have arrived at Brookhaven, and each unit is undergoing acceptance tests. An order has been placed for the construction of a 4200-v, 220-amp dc power supply for the power amplifiers.

AEC approval has been requested for the purchase of 70 Evapor-Ion high vacuum pumps, and for a contract award for the fabrication of the 240 magnet sections of the AGS vacuum chamber.

Design of control systems for the various linac and synchrotron components is continuing.

During the past year there was an increase of  $\approx 30\%$  in Department personnel. Members of the staff have presented papers at meetings of the American Physical Society and at the Naval Research Laboratory, the Congress of Vacuum Technology, Lehigh University, Stevens Institute, and the Armed Services Technical Net (MARS). A total of 46 engineers and scientists from 15 foreign countries visited the Department to discuss various aspects of accelerator design.

Details of design and construction of the Alternating Gradient Synchrotron project are described in the following paragraphs under the appropriate component headings.

### MAGNET

Early in FY 1958 a contract was awarded to the Baldwin-Lima-Hamilton Corporation for the fabrication of the 246 magnet cores for the AGS. At the same time a contract was awarded to the National Electric Coil Company for the fabrication of 986 magnet coils. Both firms submitted pilot units to Brookhaven for evaluation and approval prior to proceeding with production. The first pilot core (a long closed section, designated C-1) came very close to meeting the required mechanical tolerances. The experience obtained on this unit resulted in some changes in the assembly and welding procedure that brought the two other pilot cores (a short open core, designated B-1, and a long open core, designated A-1) well within specification. Sixteen pilot coils were submitted for approval to Brookhaven. Minor difficulties encountered with the first few units off the assembly line were easily rectified, and production was started on the balance of the order. To date, 242 coils and 47 cores have been delivered to Brookhaven.

The steel chosen by Baldwin-Lima-Hamilton for the magnet cores is an electrical grade M-36 silicon ( $\approx 1\frac{3}{4}\%$ ) sheet steel, 0.031 in. thick, from the Allegheny Ludlum Steel Corporation. Extensive permeameter measurements on a pilot order of steel resulted in the choice of a preferred normalizing process to be followed for the production or-

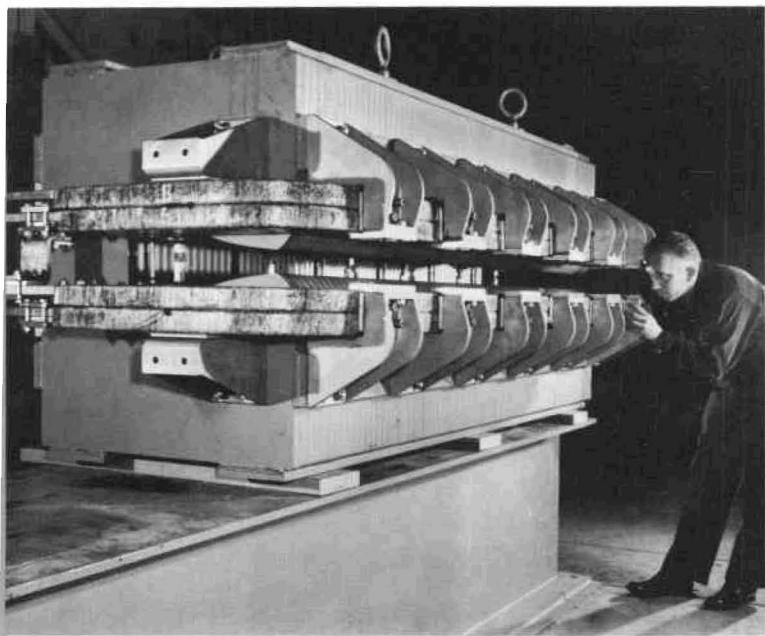


Figure 1. AGS magnet assembly. The contour of the magnetic pole tips can be seen at the front end of the magnet.

der. All the steel for the three classes of magnet units has now been processed and delivered to Baldwin-Lima-Hamilton. During the pouring, rolling, and processing of each class of steel, a Brookhaven inspector was at the Allegheny Ludlum plant to check the necessary uniformity of chemical composition and fabrication procedures. Samples of all the steels are being checked by permeameter measurements, and observations are continuing on the changes in magnetic properties to be expected with aging. At the Baldwin-Lima-Hamilton plant, the steel sheets of each class are being shuffled in such a way that the steel from a given coil of a given heat of steel is situated in the same position in every magnet of a given class. The steel for both class A (long open) and class B (short open) magnet units has now been shuffled.

Detailed mechanical measurements are being made of the fabricated magnet cores at Baldwin-Lima-Hamilton; to date all production units have met mechanical specifications. The physical lengths of the cores are uniform to an accuracy of  $\pm 0.05\%$ , and their weights are the same within the accuracy of measurement,  $\approx 0.3\%$ . The uniformity and accuracy of the pole tips are determined by a combination of precise optical and electrical measurements. Three pairs of condenser

plates are symmetrically positioned across the radial extent of the gap, three plates being located close to the upper pole and three close to the lower pole. When the capacity of an upper plate is balanced with that of its corresponding lower plate, optical measurements determine the vertical symmetry of the gap. Similar checks of the correct pole-tip spacing at given horizontal positions are made by balancing the sum of the capacities of an upper and lower pair of plates against a standard capacity. The accuracy of this instrument is about  $\pm 0.002$  in.; preliminary measurements on the magnet cores show rms deviations of the order of only 0.005 in.

The development of magnetic measuring equipment and techniques has culminated in the programs for measuring the pilot and production magnet units. Because all the pilot units were made from class A type of steel (and shuffled like class A magnets), extensive measurements were carried out only on the pilot unit A-1 (long open). However, checks of the remanent field in the C-1 pilot unit (long closed) showed that, because of geometry differences alone, these remanent fields differed from those in A-1 by only  $\frac{3}{4}$  gauss. Azimuthal variations in remanent field tracked closely, which showed the efficacy of the shuffling procedure. Over the range 500 to 10,000 gauss, point measurements of the fields and gradients in the A-1 unit agreed closely with computed values, and the gradient-to-field ratio was constant over the full 6-in. radial aperture within the accuracy of measurement,  $\approx 0.2\%$ . The remanent fields in A-1 were somewhat lower than expected,  $\approx 14$  gauss.

To measure the integrated effects on the proton beam of a full magnet unit, three 10-ft-long search coils have been designed and built. With these coils it was found that along the aperture centerline the magnetic fields off the ends of the magnets contribute an extra "magnetic length" of field of 4.1 in. over the physical length, in close agreement with computations from point measurements made on some of the magnet models. This "magnetic length" remains constant to  $\approx 0.2\%$  from injection field values to about 10,000 gauss and shrinks by only about 1% at the top central field of 13,000 gauss. As expected, the end effects in the magnet also give rise to a small radial second derivative in the integrated fields; this amounts to a rate of change with radius in the integrated gradient-to-field ratio of  $\approx \frac{1}{3}\%/\text{in.}$  over the magnetic aperture for fields below saturation values. At the

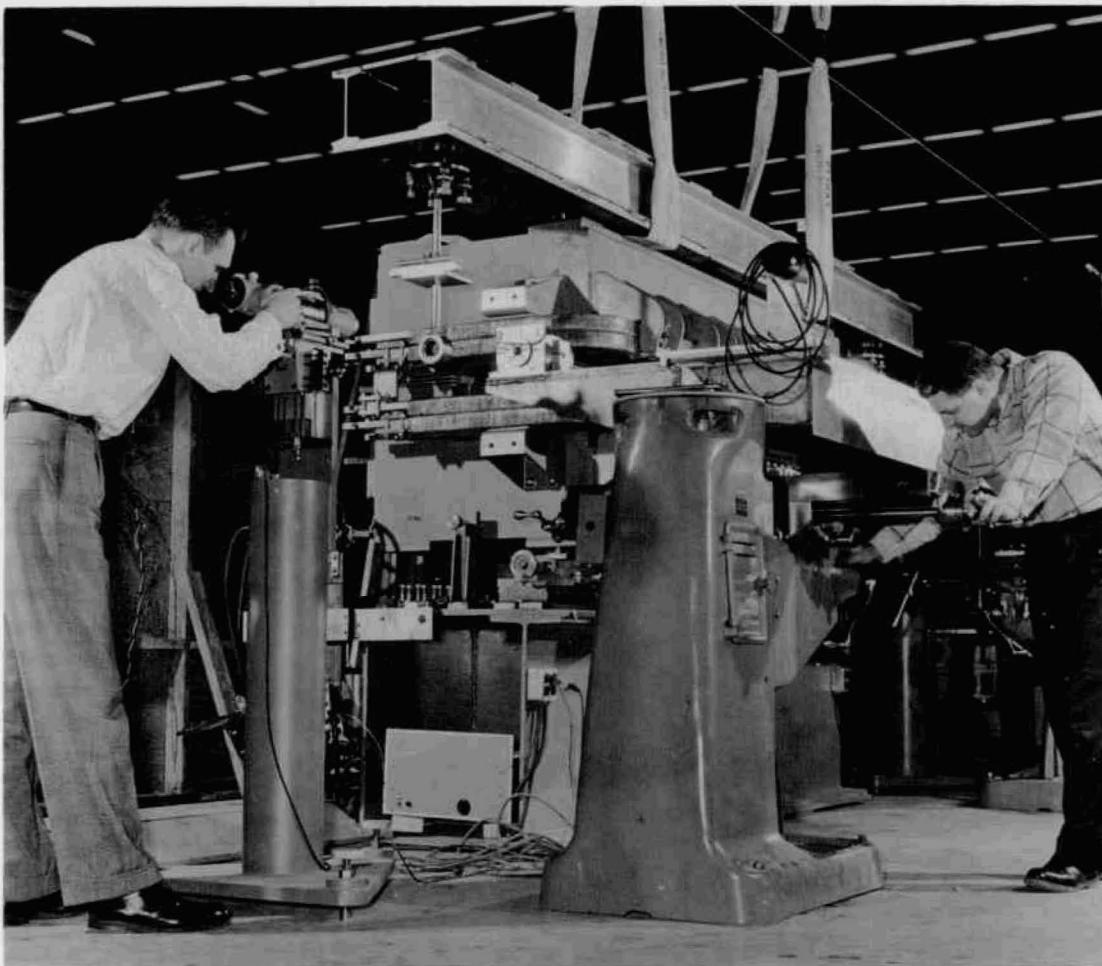


Figure 2. Testing one of the AGS magnet assemblies.

top fields, the integrated gradient-to-field ratio is reduced (because of saturation effects) by  $\approx 4\%$  at the aperture centerline; its rate of change with radius, across the aperture, is  $\approx 3\%/\text{in}$ . These amounts are somewhat lower than those obtained in the magnet models; the improvement is due to final design changes and to a good packing factor for the steel. To establish standards of accuracy and procedure for production testing, very detailed magnetic and mechanical measurements are being made at BNL on the first few production magnet units.

In order to make small corrections in the magnetic field gradients, particularly at saturation, two quadrupole magnets will be inserted in each superperiod of the magnet arrangement; one quadrupole corrects the vertical focusing and the other the horizontal focusing. From the results of

magnetic measurements on the models and the pilot unit, the maximum change in gradient needed is  $\approx 5\%$ ; this results in maximum pole-tip fields of about 7500 gauss in 6-in.-diam quadrupoles. Three sextupoles will be inserted in each superperiod for correcting second derivatives in the magnetic field; two of these make corrections in the horizontal direction and one in the vertical direction. Again, the maximum correction is needed at the top fields, and magnetic measurements indicate maximum pole-tip fields in the sextupoles of  $\approx 6000$  gauss. Detailed orbit calculations have determined the optimum location for these multipoles. Several quarter-scale models of each type have been built and measured, and the results, together with some analytical studies, have provided enough information on their power requirements so that preliminary design can be

started for the power supplies and their controls. Mechanical design of the multipoles and their coil configurations has also begun.

Eddy currents in the Inconel vacuum chamber not only produce small changes in the magnetic field shape but also cause a reduction in the guide field itself. Variations in the resistivity and thickness of the chamber walls can produce sufficient disturbance in the particle orbits to be troublesome; to reduce these effects, a saturable inductor will be introduced into the magnet circuit to reduce the rate of rise of the magnetic field during the first 100 msec of the cycle. Models of such an inductor have been built and tested, and further analysis has resulted in a final design to be ordered shortly. This inductor will also form part of a filter circuit, probably a shunt-type regulator, that will reduce the output ripple from the main magnet power supply. To reduce initial transient effects, each magnet has a resistance of  $\approx 50$  ohms, connected in shunt, and these are on order.

Even with the reduced rate of rise, the eddy currents in the vacuum chamber produce unwanted changes in the magnetic field shape. Computations and a series of measurements carried out on Magnet Model No. 7 showed that a few windings, each carrying about 1 amp and distributed non-uniformly across the surface of the chamber, would provide adequate compensation.

The Westinghouse Electric Corporation has delivered to the Laboratory most of the switchgear, all the transformers, and all ignitrons and auxiliary equipment for the main magnet power supply. Fabrication of the prime mover and alternator has been delayed, pending the incorporation of some design changes which are a direct result of information obtained from the failure of the Bevatron magnet power supply at Berkeley.

The over-all control scheme for the power supply was submitted to the Laboratory for approval. The staff devoted considerable time and effort to circuit modifications in order to improve the operation and reliability of the system. The circuit details were worked out in conference with the vendor's engineering representatives.

Surveying of the synchrotron ring is a continuous program. Initially, two precise leveling circuits were run connecting marks on the pile caps in the magnet enclosure to existing permanent bench marks on site. After verification, the Stone and Webster Engineering Corporation's data on the location of prick marks on each of the 24 primary

survey monuments were used in setting the magnet girders. A complete closure survey was undertaken of the 24 survey monuments (angles and distances) to provide the basic information for magnet location. During the erection of the high density shielding block in the target building, the closure procedure was repeated. It was found that all but the three monuments in the target building are within 0.005 in. of the specified distances, and all angles are within one second of  $165^\circ$  (inside angle). Setting of the secondary monuments located between girders has started.

### LINEAR ACCELERATOR

A contract was awarded to the Lukeweld Division of the Lukens Steel Company for the fabrication of the eleven 10-ft-long tank sections of the linear accelerator. This firm has completed the forming and welding of all units and has shipped them to the Cleveland Diesel Corporation for final machining to size. Section No. 1 was completed in June 1958 (after some reworking due to vacuum leaks) and was delivered to the Laboratory. Sections No. 2 and No. 3 are completed and are awaiting final inspection and approval before shipment.

Early in 1958 it became obvious that the development of a suitable klystron for the linac rf power source was too slow to be compatible with the over-all construction schedule. Consequently, it was decided to drop the klystron program and design the rf system around the immediately available French-Thomson Houston TH470 triodes. In the final stage of the power amplifier, the output of two TH470's will be combined in a waveguide hybrid junction. Two TH470's will form the chain to drive the final stage. French-Thomson Houston has agreed to supply the tubes, cavities, and auxiliary equipment on a schedule consistent with machine requirements.

Production has started on the 124 drift tubes required for the linac. The special castings have been received, and rough machining of the bodies has started. The first brazing operation and contouring of 9 drift tubes have been completed. The final brazing procedure on this component is awaiting completion of arrangements at the hydrogen furnace facility leased for the operation.

When the Cockcroft-Walton preaccelerator was delivered to the site it was erected in the service building, where tests included operation at its

rated 800 kv. Subsequently it was moved to its final location in the linac building. Stiffening members have been added to the filter stack, and installation and fitting of equipment in a new filter terminal has started. A Cosmotron ion source will be used initially. The basic design of the ion source focusing system has been completed, and a theoretical analysis indicates that the proposed arrangement should be able to handle  $\approx 50$  ma of beam current.

Some difficulties have been encountered with the first linac focusing quadrupoles delivered for approval. A moderate amount of redesign is needed to improve the voltage standoff characteristics of these units, and the appropriate steps are being taken.

Design studies made of the AGS injection system include an array of bending and focusing lenses to steer the beam and match the emittance of the linac to the admittance of the synchrotron. The use of a beam chopper, debuncher, and momentum analyzer, as well as beam observation equipment, has been proposed, and preliminary designs have been completed.

The linac "high power" model has proved to be an extremely useful tool in evaluating the performance of the auxiliary equipment to be used on the linear accelerator. For instance, probes for measurement of the linac radio-frequency fields have been tested and calibrated in this system, and the behavior of various solders and plastics under high vacuum, high field conditions has been determined.

#### RADIO-FREQUENCY

During the past year, considerable effort has gone into the improvement of the design and reliability of the over-all rf system. A prototype of the final stage of the power amplifier has been built and tested. This unit is the forerunner of 12 such assemblies to be located around the magnet enclosure. The original test model power amplifier was modified to a cathode follower and used to drive the prototype through 1600 ft of coaxial cable, which simulated the proposed final arrangement. Full voltage with little harmonic distortion was achieved over the desired frequency range of 1.4 to 4.5 Mc/sec. Specifications covering the fabrication of the 12 final stages of the power amplifier, plus 1 spare unit, are being prepared to accompany bid solicitations.

Experimental work continues on the low level rf system. (The operating principle of this system was described in the previous annual report.) Experimental circuits of the multiple heterodyning equipment using an 0.45-Mc/sec carrier have been constructed and successfully tested. Self-tracking amplifiers have been designed for use in the low level system. These amplifiers stay resonantly tuned over the desired frequency range, and the output signal is free of harmonic distortion with a minimum amount of differential phase shift.

The Philips Company of Eindhoven, The Netherlands, has shipped to Brookhaven 860 ferrite rings for the accelerating cavities. Of this total, only 29 units failed to meet BNL specifications. Nineteen of these rejected units have high *Q* values, but do not meet the permeability tolerance of 350 at zero rf flux density. The remainder exhibited low *Q* values at 100 gauss rf flux density.

Proposals were received and evaluated, and a contract was awarded to the Moloney Electric



Figure 3. Inside the magnet tunnel looking toward the Cockcroft-Walton assembly. A portion of the platform on which the magnet will be mounted is visible. The upright cylinder at left front is one of the survey monuments.

Company for the fabrication of the 4200-v, 220-amp dc power amplifier power supply. This supply will provide dc power for the 12 final stages of the rf power amplifiers as well as the single high level remote driver.

Work has started on the basic design of the AGS timing system.

### VACUUM

During the past year experience was gained in the operation of the Evapor-Ion pumps, and some design modifications were made to increase their reliability, with the result that their use throughout the AGS seems feasible. AEC approval has been requested for the purchase of 70 units to meet the requirements of the synchrotron and linac.

AEC approval also has been requested for the award of a contract for the fabrication of 240 magnet sections of the AGS vacuum chamber. The chamber will have a  $3\frac{1}{4} \times 7$ -in. cross section and is to be made of 0.78-in.-thick Inconel-X, the end flanges being stainless steel. Correcting windings will be mounted on the vacuum chamber to take care of eddy currents and remanent field effects. Since only limited space is available for these windings, the use of a recently developed tape cable appears attractive. This material consists of copper ribbons embedded between two layers of polyester insulation (total thickness, 0.008 in.). The use of two 3-in.-wide ribbons on top of the chamber and two more on the bottom makes available a total of 58 separate conductors, each capable of carrying 1 amp.

Work continued on the development of a ceramic-to-copper seal for the rf accelerating cavities. Because of the large pieces involved, standard processes could not be used. A promising technique utilizing epoxy resin as the binding material has been worked out. A test bond has proved to be vacuum tight and to have no undesirable properties as an rf insulating material.

### CONTROLS

During the past year much effort was devoted to the development of the water cooling and air conditioning systems for the Alternating Gradient Synchrotron. This included the design of a scheme for controlling the temperature and differential pressure in both the main magnet and experi-

mental magnet cooling loops. The development of the scheme required detailed analysis of system operation, flow and pressure characteristics, process loops and feedback characteristics, and finally the selection and specification of appropriate control valves.

An automatic sequencing circuit has been designed for remote and unattended operation of the three deep-well supply pumps. A board has been designed to centralize the controls for all water loops and for the main air conditioning systems. It will contain various pneumatic and electric indicators, recorders and controllers, malfunction annunciators, and system interlocks as required for the effective operation of all these systems. Delivery of the control components is scheduled for August 1958; meanwhile, the board itself is being fabricated.

A detailed inspection of the air conditioning systems supplied by the building contractor has been made, and Department engineers have worked with the contractor to put the system into acceptable operating condition.

AGS requirements for an intercommunications system were prepared, and discussions were held with possible vendors. However, cost considerations made it advisable to have this equipment designed and installed by BNL personnel. A system has been devised which permits operation of up to 10 conference circuits (with 6 telephones each) among the 125 stations located throughout the synchrotron complex. Facilities for selecting the desired conference circuit will be provided at each station. In an attempt to get an acceptable public address system, various acoustical tests have been performed in the magnet enclosure, where the acoustics are very poor. An arrangement has been devised which gives marginal performance at present, and acoustical tile may be required in the final installation.

Considerable time has been spent on the main magnet power supply controls. Much time has also been spent on the linac controls, with emphasis on the Cockcroft-Walton set, the rf system, and the linac control console.

Design of the controls is nearly complete for the Evapor-Ion pumps in particular and the vacuum system in general. The system design for the pumps has required a detailed analysis and measurement program of their operating and electrical characteristics. It is planned to use ionization gauges for measuring vacuums. The design and

specifications for magnetic amplifiers for these gauges have been prepared, and prototype amplifiers procured.

### STRUCTURES

During the past year the major portion of the AGS building complex was completed, and the Department has taken occupancy of the linac building, target building, and magnet enclosure.

Work has started on the completion of the synchrotron power room. The large foundation for the main magnet power supply has been erected. The L.A. Wenger Company is proceeding with the necessary excavation and trench forming work in this area.

In October 1957 the pouring of the high density concrete shielding blocks was started. The work was delayed by both strikes and adverse weather, but by April 1958 all blocks had been fabricated and erected in the target building.

The main water supply for the AGS will be provided by three wells capable of delivering up to 2800 gal/min. An analysis of the water shows that the iron content ranges from 12 to 22 ppm, which is higher than that predicted on the basis of data obtained from test wells and already existing wells in this area of Long Island. Continuous pumping on the system should tend to reduce the iron content to an acceptable level.

A warehouse has been constructed adjacent to the target building to facilitate receiving, handling, and storage of machine equipment.

## Instrumentation

Substantial progress has been made on the high-capacity digital computer started in 1956. Basic design was completed this spring; power supplies and input-output equipment have been delivered, and about one-third of the electronic circuits have been built.

Digital data handling, which involves electronic computer techniques, is rapidly being applied in research laboratories. Consequently the efforts of several members of the Division are concentrated on the design and construction of instrumentation for this purpose. Included are multichannel pulse height sorters; multichannel pulse time sorters, both fast and slow; automatic recording devices for meteorological data; neutron spectrometer data; and cloud chamber track analysis.

The ultrafast oscilloscope developed here has been used to study the characteristics of photomultiplier tubes and transistors in the range of a billionth of a second.

Several members of this Division worked with members of the Medical Department to prepare and operate instrumentation for a study of the whole-body activity of the Marshall Islanders exposed to radioactive fallout during the 1954 Pacific weapons test series. This involved acquiring large scintillation detectors and multichannel analyzers, testing the equipment in the steel-shielded room procured for this purpose, and making sure that the equipment worked properly after it was shipped to the Pacific. Because data and electronic equipment had to be jettisoned at sea on the return flight, it was necessary to repeat the entire operation.

Dr. Martin Graham, in charge of the computer for the first year, transferred to Rice Institute, Houston, Tex., to build a similar computer there. Mr. Robert Spinrad headed the computer group this past year. The group, with the addition of 2 engineers and 3 technicians, now has 11 members. Two scientists and two technicians joined the Instrumentation staff. The Division now has a total of 52 members. This is the first appreciable expansion of the Division since the start of the Laboratory. The increase was required to complete the computer in a reasonable time and to develop the

increasingly complex apparatus needed by other departments.

### DIGITAL COMPUTER

A general purpose digital computer consists of three sections, arithmetic, memory, and control. The arithmetic section was designed last year, but the pulse transformers appeared to be unreliable. Toroidal core transformers were substituted and arranged so that any desired number of output connections could easily be attached. Construction of the arithmetic section is nearly complete. Considerable difficulty has been experienced in obtaining satisfactory barrier grid storage tubes for use in the memory. Usable tubes will be available this fall, but it is too early to predict how many spots they will store or the ultimate memory size. Much time was devoted to test equipment for storage tubes and to experimental memory sections. Interference between the writing pulses and the read signals, a common problem in this type of memory, has been completely eliminated. Amplifiers and pulse control circuits for use with the memory tubes were designed with radio tubes, and then redesigned with transistors. The Applied Mathematics Division worked out the order code and logical design of the control section. Circuits have been designed for the control section, and construction has started. A 40-kw motor generator set and the plate power supplies were delivered, installed, and tested. A high speed printer, paper tape input-output devices, and magnetic tape auxiliary storage units were procured. Work has started on attaching these machines to the computer and on construction of the control console.

### DIGITAL DATA HANDLING

Automation will be of great assistance to research scientists. In some cases it may be used to record data automatically, and at a much faster rate than is possible manually. In other cases the automatic data processing equipment can sort or correlate the data, and thus perform tasks which are otherwise very laborious. It is desirable in

many cases to transform electrical signals into numbers which may be fed into high speed digital computers for further analysis. Three major projects of this type will be described briefly here.

Detailed study of the behavior of the atmosphere near the surface of the earth under different atmospheric conditions is of interest for understanding such problems as the smog associated with industrial gaseous wastes and the stresses which will be imposed on the large radar-type antennae being built for radio astronomy. The BNL meteorological tower has wind and temperature measuring instruments at several levels. Automatic data handling equipment was built which converts the wind angle, velocity, and temperature readings to numbers and punches a complete set of data on paper tape in a fraction of a second. It will record 6000 complete sets of data an hour.

In the last few years 100-channel pulse height analyzers have been developed which are extremely useful in research and industrial applications of scintillation radiation detectors. In many cases radioisotopes emit two or more radiations simultaneously which may be detected in separate detectors. It would be of value to be able to analyze both radiations and record the spectrum of pulses occurring in one detector in coincidence with each value of pulse height occurring in the second detector. As a start in this direction an instrument has been designed at BNL which has 32 analyzers, each with 96 channels, arranged to record the spectra in one detector for each of 32 values of pulse height in a second detector. This instrument is completely transistorized with the exception of the display circuit. A magnetic drum is used for storing the accumulated data. The instrument is in fact a special purpose digital computer. It will have a variety of displays on a cathode-ray tube and provision for printing the data. It uses about 1500 transistors.

The third large project of this type is construction of a 1024-channel time analyzer similar to the one which was described in the 1955 Annual Report and is now in use with the fast neutron chopper at the reactor.

#### TRANSISTOR CIRCUITS

Transistors will perform most of the functions for which radio tubes have been used in the past. Transistor circuits often require much less power, take little space, and may be more reliable. Since

power and size are not of major concern in the laboratory, there is little reason to redesign research equipment which functions properly. Either tubes or transistors will be used for new circuits, depending on the particular application. Often transistors have advantages besides small size and low power. For example, they make excellent high speed electronic switches; power transistors may be used to control large currents at low voltages; and fast transistors which hold great promise for fast pulse circuits are becoming available. They were used in the  $32 \times 96$ -channel pulse analyzer described above primarily because of their good switching characteristics. Some conventional circuits such as a pulse preamplifier have been transistorized for use in confined locations. A fast pulse amplifier - electronic counter unit which will count up to 4 Mc/sec was redesigned with transistors, and considerably better performance was achieved at lower cost. Electronic regulation of the current in magnets is often required in the laboratory. Power transistors were used to build two circuits for regulating 0 to 150 amp at 36 v. The ultrafast oscilloscope developed here has been used to study the performance of new transistors in the time range of a billionth of a second so that transistors may be used in experiments in this short time range.

#### MISCELLANEOUS DEVICES

A device was built to measure the water level in a critical assembly. The amount of water determines the neutron multiplication or "criticality." The water level is measured to an accuracy of 0.005 in. out of 4 ft. A simple, precise analogue computer was built to enable the Nuclear Engineering Department to record the percent concentration of magnesium in a reactor solution, which required taking the ratio between certain voltages and recording the answer on logarithmic paper. The Division also assisted Nuclear Engineering personnel in developing a magnetic method of continuously measuring the wall thickness of an iron pipe under erosion. A combination tube and transistor instrument was designed for reading pocket radiation dosimeters.

#### METEOROLOGY

The work of the Meteorology Group continues to be divided between research and service func-

tions, although the emphasis at present is pre-ponderantly on research and development. A few new projects have been undertaken during the past year. One of these is an investigation of the deposition of particulates on the ground and vegetation surfaces.

### Deposition

Past experience with field experiments in diffusion suggested that this study would include several painstaking developments before useful field data could be obtained. The first such development, the production of suitable tracer particles, has been virtually completed. The requirements for an acceptable tracer are extremely severe. Easily dispersible, chemically inert particles are needed whose size, or at least size range, can be controlled. The use of a radioactive tracer is almost mandatory in this type of work, since the field experiments must be repeated in the same location at relatively frequent intervals. Radioactivity seems to be the one property that will assure reproducibility without cross contamination from one experiment to the next. Montmorillonite clay particles containing  $\text{La}^{139}$  possess all the qualities mentioned. They can be fused into perfect spheres ranging in size from 0.5 to 25  $\mu$  and then separated into groups of the desired sizes. Figure 1 shows a group of typical particles prior to sizing. Neutron irradiation in the reactor converts some of the  $\text{La}^{139}$  to radioactive  $\text{La}^{140}$ .

The next development involves the dispersal of relatively small numbers of particles at steady rates in such a way that no serious agglomeration or other alteration of the particle size occurs. Calculations indicate that emission rates of the order of not more than a few mg/hr are required. This probably can best be achieved by introducing the clay spheres into a relatively large volume of a volatile fluid such as alcohol and subsequently dispersing the fluid through a fog nozzle.

### Diffusion

Studies of short-range diffusion (0 to 10 miles from the source) have been continued with the objective of establishing firm relationships between meteorological variables and field concentrations observed from a specific diffusion source. It has been recognized for a considerable time that the diffusion process from a continuous point source is strongly dependent on time. This particular aspect of the problem has been given detailed con-

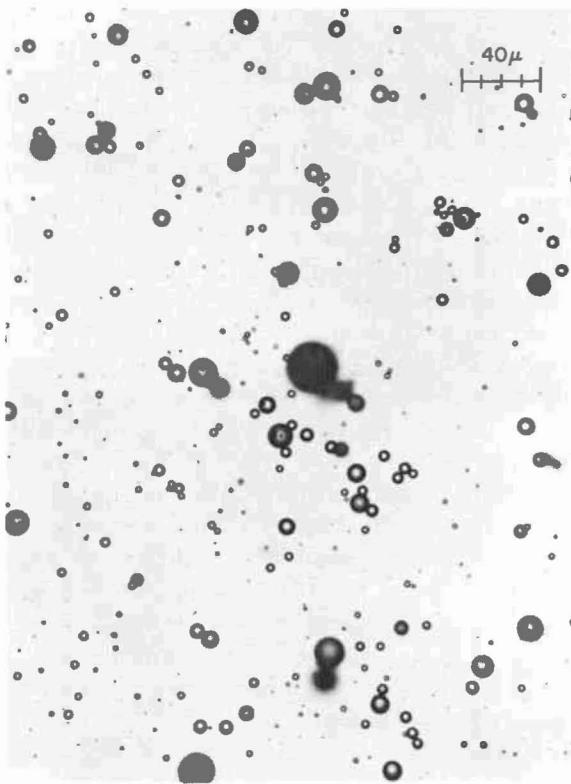


Figure 1. Clay spheres prior to sizing. The spheres are shown on a microscope slide as they appear after fusing in the oxyacetylene flame. In this particular group, the particles range in size from a small fraction of a micron to  $\approx 20 \mu$ .

sideration. Analysis of existing data indicates that the peak concentrations observed over successively longer time periods in any given diffusion test are related in a mathematically reliable fashion, and that the parameters of the equations defining these peak-mean relationships are strongly associated with the Brookhaven wind gustiness classification. It is not yet possible to define precisely the physical relation between these parameters. This matter will be investigated further during the summer of 1958. The study should assist materially in establishing empirically the peak concentrations to be expected in various sampling times and under differing meteorological conditions.

A new study in the Brookhaven meteorological program this year is that of the long-range diffusion of smokes and gases. So far, this has consisted mainly of qualitative exploration and the development of instruments. For studies of diffusion at distances of 50 miles or more from the source, a very dense network of sampling stations in both

the vertical and horizontal planes would be preferable, but establishment of such a network would involve great expense. The alternative is a light aircraft to serve as a highly mobile sampling probe as well as the vehicle for appropriate meteorological instruments. Since visual experiments had shown that oil-fog plumes such as those used in past studies at BNL will serve as suitable tracers at least to distances of 50 miles, work was begun on the development of an airborne sampling unit for measuring oil-fog. While the light-scattering principle embodied in earlier successful instruments can be utilized in this unit, many operating characteristics have had to be changed. The device now nearing completion is expected to sample and record oil-fog concentrations at speeds up to 75 mph.

### Instrumentation

An automatic data recording system, developed in collaboration with the Instrumentation Division, has been installed in the meteorology building. Figure 2 shows the assembly with the tape reels in the foreground and the servomechanisms and analyzer to the rear. At present, the instrument is capable of recording wind direction (either vertical or horizontal), wind speed, and temperature at accuracies surpassing those available in the usual strip chart recorders and with a scanning cycle of 0.6 sec. In the future it will be able to record any variable which may be translated in the form of a rotation or a voltage and will serve as the primary recording device for all meteorological data at this site.

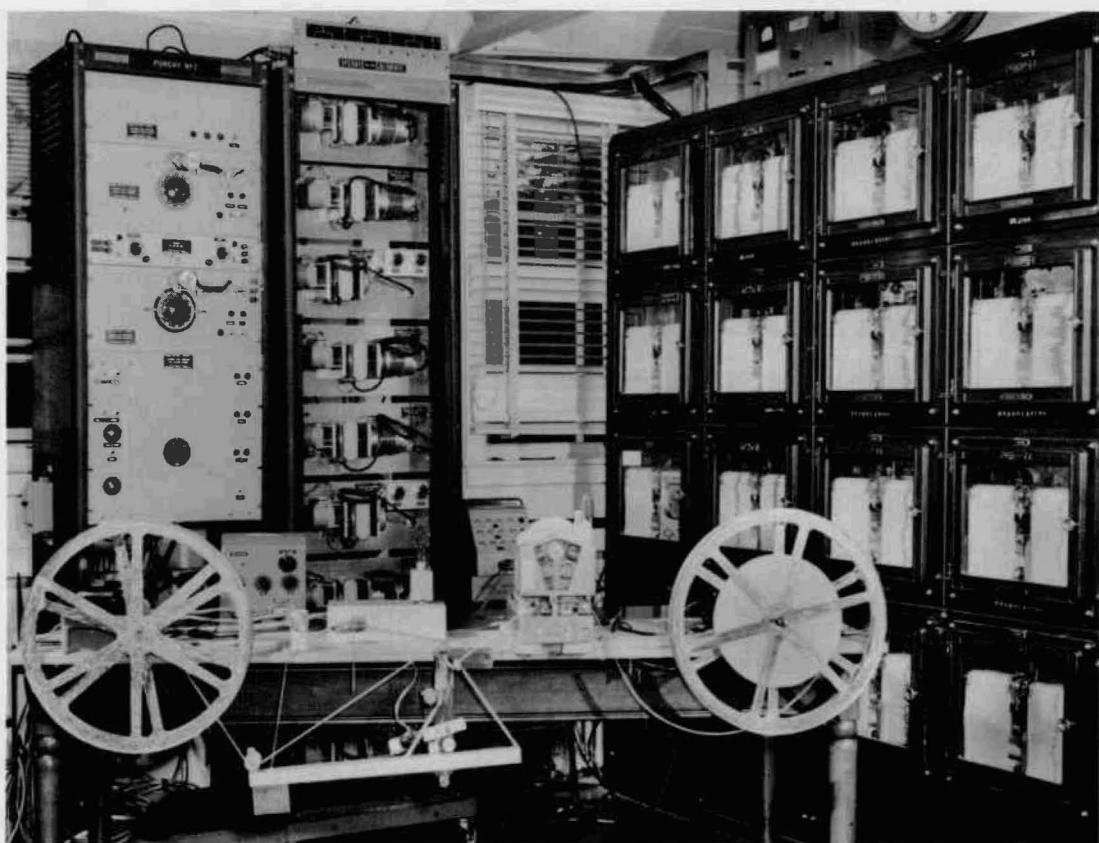


Figure 2. Automatic data recorder. The photograph shows the complete equipment for automatic tape recording of meteorological data. The 18-in. reels and take-up assembly in the foreground will record continuously for 2 hr at a cycling rate of 0.6 sec. The lever and roller take-up mechanism is required to keep uniform tension on the relatively large tape reels. The processing and calibrating equipment is seen in the left background, and the servomechanisms are in the adjacent vertical bank. The panel on the right is the original recording system utilized since the inception of the meteorological program.

A new type of wind direction transmitter has been developed; it utilizes relatively inexpensive parts to provide a straightforward, unambiguous record on any standard strip chart recorder. Perhaps the most useful aspect of the new wind vane is that it can be coupled to several individual recorders at various locations on the site by the direct use of two telephone leads and without the need of repeating or amplifying equipment. The first of these wind vanes has been transmitting data from the meteorology tower to the reactor over more than a mile of telephone line without difficulty for some six months.

#### **Related Meteorological Projects**

An agreement has been concluded with the U. S. Army Signal Corps for an investigation of

the variation of low level wind with height. The problem is summarized in the question, "With what instrumentation and knowledge can a single observer in the field predict the vectorial change of wind with height, and what is the probable accuracy of this estimate?" This implies not only an investigation of existing data but a new series of unusually accurate measurements, all to be related to some reference wind measurement and classified in the simplest meteorological terms possible. Work on this project has so far covered only existing data. The measurements with the improved order of accuracy are awaiting the completion of a small wind tunnel and the delivery of new propellers for the existing wind instruments. Other new equipment, including a device for measuring the net radiation exchange between the earth and the atmosphere, is on order.

## Applied Mathematics

The Applied Mathematics Division, organized a year and a half ago, is still in its early period of growth. Within the past year its staff has been increased to 21 persons, of whom 9 are mathematicians and physicists and 12 are programmers or other technical specialists. Further expansion is planned for the coming year.

### COMPUTER RESEARCH AND DEVELOPMENT

Merlin, the high speed digital computer under construction by the Instrumentation Division, occupies a central role in the activities and planning of this Division. Although this computer is patterned after the Maniac II computer, now in operation at Los Alamos, a number of special features have been incorporated in its design. Four high speed access registers have been added to the arithmetic section, and, principally because of this feature, the computer instruction word has been modified to a combined 1, 2, and 3 address form. An irregular fetching of instructions from memory has been introduced in order to minimize the time lost in operating the machine on the basis of one instruction per word in contrast to two instructions per word for Maniac II. A study of the logical structure of the extract function has led to the incorporation of several special logical operations. Also, a special feature has been introduced which allows the marking of arithmetic numbers by tag bits which are automatically sensed by the machine. These modifications, among others, have necessitated major redesign of the logic of the control section by members of this Division working in close collaboration with the Instrumentation Division.

A broad study of various programming systems developed for other computers was undertaken before finalization of the specifications for the system to be developed for Merlin. A proposal for a system for this machine is now under consideration, and intensive development in this direction will be undertaken during the coming year.

Several studies in numerical analysis were begun, and a new technique for studying various

interphase phenomena was investigated. A technique suggested by this method is now being studied as a possible approach to the direct numerical treatment of detonation and deflagration processes.

During the year a seminar on magnetohydrodynamics was included in the activities of the Division.

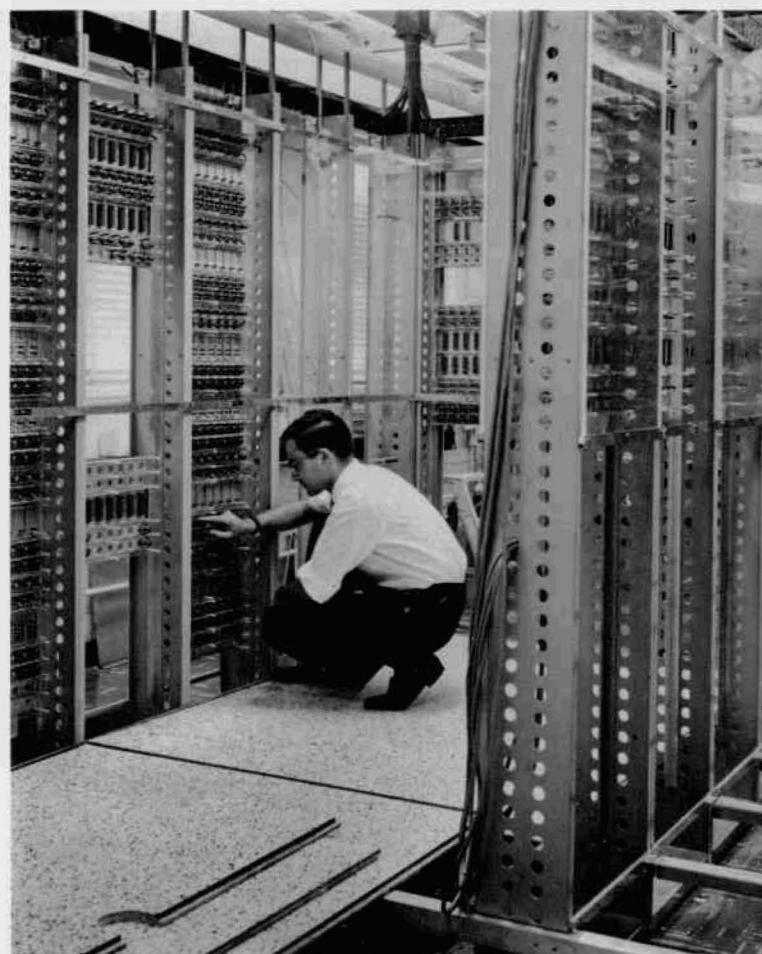


Figure 1. Inspecting a completed portion of the arithmetic section of the digital computer under construction. In this view of the computer's interior the control is on the right, and the arithmetic section stretches off to the left. The elevated flooring permits the flow of cooling air into the vertical rack structure.

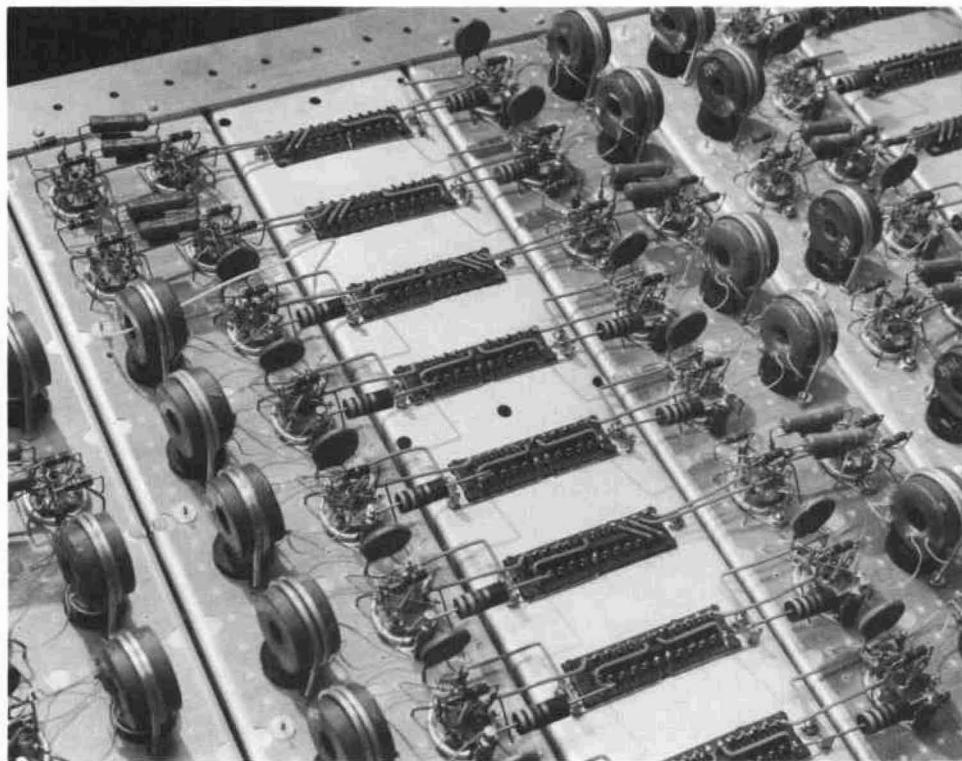


Figure 2. Part of the control section of the digital computer under construction. Shown here are 16 individual control circuits prior to cross-connection with 400 of their neighbors. The control section directs the movement of information between the various parts of the machine and controls communication with the human operator.

## Chemistry

Research activities in the Chemistry Department have continued to follow the general pattern set forth in previous annual reports. In the field of nuclear chemistry, work on high energy nuclear reactions has been temporarily halted by the breakdown of the Cosmotron. Analysis has continued of tracks in nuclear emulsions exposed before the breakdown occurred, with the emphasis on angular distribution of alpha-particle tracks. Data from Monte Carlo calculations on nuclear cascade processes have been used to investigate the competition between fission and evaporation from excited heavy nuclei. Studies of the half-lives and characteristic radiations of  $I^{126}$ ,  $Tc^{97}$ ,  $Ru^{97}$ ,  $Ge^{65}$ ,  $Gd^{146}$ , and  $Ba^{129}$  have been carried out.

The use of the mass spectrometer in nuclear chemistry has increased during the past year. With this technique, the production of cesium in uranium and the production of rare gases in iron have been investigated. The measured cross sections for rare gas production have been combined with determinations of rare gas contents of iron meteorites in order to determine cosmic-ray exposure ages of the meteorites. An aspect of mass spectrometry new to this Department is the high precision determination of mass differences of isotopes involved in beta decay.

Studies on chemical effects of nuclear transformations in both inorganic and organic systems have been continued. Thermal annealing effects in inorganic solids have been discussed in terms of various physical models.  $C^{14}$  and  $C^{11}$  recoil experiments continue to show the importance of chemical rather than physical factors in the determination of products. Organic compounds which have been investigated are aniline, benzene, and pentane.

Extensive use has also been made of isotope exchange reactions as a tool for studies of reaction mechanisms. Certain steric factors of 1,2,3-trimethoxybenzene have been evaluated by this technique. Similar methods were used to show that a new Brønsted acid is formed when stannic chloride is dissolved in acetic acid. The rate of exchange of aromatic hydrogen in toluene with tritium-labeled sulfuric acid was found to be the same as that for toluene- $\alpha$ - $d_3$ . This result implies that

secondary isotope effects are not always connected with hyperconjugation. The effect of ion pair formation on the reactivity of bromide ion has been studied by measurements of the rate of exchange of radiobromine between various compounds.

The radiation chemistry of aqueous solutions has been considerably clarified by the recognition that two types of hydrogen atom are involved in such systems. One of these is produced by the decomposition of water by radiation, while the other is formed by free radical oxidation of a hydrogen molecule. Work on the radiolysis of pentane adsorbed on solids has shown that certain modes of decomposition are affected more than others by the adsorption process.

A new theorem concerning vibrational frequencies of a homologous series of isotopic molecules has been developed. This theorem, the second-order sum rule, facilitates the analysis of molecular spectra and extends the limits of justification of the rule of the geometric mean. In other theoretical work, certain temperature independent isotope effects in linear molecules were found to depend solely on molecular weights. This result has been applied to both equilibrium and kinetic properties of isotopic molecules.

Neutron activation analysis for  $A^{40}$  and  $K^{41}$  in iron meteorites has been used to estimate ages of the meteorites. These ages were significantly higher than those of stony meteorites, a result of considerable cosmological interest. In another application of radiochemical techniques, radioactive silver has been used to study ion exchange equilibria on single resin beads. A surprising lack of uniformity among the beads is found.

Several aspects of structural chemistry have been investigated. As in previous years, the largest part of this work is concerned with neutron diffraction investigations. One of the advantages of this technique, as compared with x-ray diffraction, is the ease with which hydrogen atoms can be located. Dimethylglyoxime and sulfamic acid were studied from this point of view. In connection with such single-crystal work, use is being made of the New York University Computing Facility, and a library of programs for routine crystallographic calculations has been accumulated. Neutron dif-

fraction has also been used to show that the second-order phase transition in magnetite ( $\text{Fe}_3\text{O}_4$ ) involves an ordering of the ferrous and ferric ions. The structure and magnetic properties of chalcopyrite ( $\text{CuFeS}_2$ ) have also been investigated by neutron diffraction. The Heisenberg-Dirac model for ferromagnetism and antiferromagnetism has been extended, and the results indicate that the model is useful for quantitative predictions of high temperature magnetic properties.

A quite different aspect of structural chemistry is presented by the study of spectroscopic properties of proteins at low temperatures. Solvents have been found in which proteins are soluble at low temperatures, and which form glassy solids at liquid nitrogen temperatures. This has made it possible to obtain fluorescence and phosphorescence spectra of dissolved proteins.

## NUCLEAR CHEMISTRY

### Nuclear Reactions

Work on high energy nuclear reactions was severely curtailed by the breakdown of the Cosmotron on November 5, 1957. As a consequence, several nuclear chemists have diverted their efforts to studies of low energy reactions. Several investigations of excitation functions of alpha-particle induced reactions, including  $(\alpha, p)$ ,  $(\alpha, n)$ ,  $(\alpha, 2n)$ ,  $(\alpha, pn)$ ,  $(\alpha, 2p)$ ,  $(\alpha, 3n)$ ,  $(\alpha, p2n)$ , and  $(\alpha, an)$  processes, are in progress at the 60-in. cyclotron.

One of the high energy reaction studies completed before the Cosmotron breakdown was the measurement of the absolute cross section of the  $\text{C}^{12}(p, pn)\text{C}^{11}$  reaction with 2 and 3-Bev protons. The measurements, carried out in the external pencil beam of the Cosmotron, involved direct counting of the proton beam intensity with counter telescopes and determination, by low level counting techniques, of the  $\text{C}^{11}$  activity induced by the beam in a plastic scintillator. The cross sections were found to be  $26.0 \pm 0.9$  mb and  $26.6 \pm 1.0$  mb, respectively, at 2.0 and 3.0 Bev. These measurements establish a much more accurate cross section scale in this energy region than was hitherto available.

Nuclear emulsion studies of fragments emitted in high energy nuclear reactions have been continued, since some emulsions exposed to beams of 1.0, 2.0 and 3.0-Bev protons were available at the time of the Cosmotron failure. The  $200\text{-}\mu$ -thick Ilford D-1 emulsions used are relatively insensitive and

register alpha-particles with energies up to  $\approx 50$  Mev and fragments heavier than alpha-particles. Techniques have been developed for distinguishing alpha-particles, fragments with  $2 < Z \leq 6$ , and still heavier fragments. Three types of stars produced in silver and bromine nuclei have been investigated: 1) those in which, in addition to the recoil nucleus, only alpha-particles are observed, 2) those in which fragments with  $2 < Z \leq 6$  are also found, and 3) events in which two heavy fragments (with  $Z > 6$ ) are observed.

The angular distributions and energy spectra of the alpha-particles have been studied. If, as is likely, the alpha-particles originate chiefly in evaporation processes and are emitted isotropically in the system of the moving nucleus, their angular distribution is a measure of the momentum of the moving nucleus. At 3-Bev bombarding energy, the ratio of alpha-particles emitted in the forward hemisphere (with respect to the beam direction) to those emitted in the backward hemisphere is 1.16, 1.3, and 1.4, respectively, for events of types 1, 2, and 3. This may be taken as an indication of larger energy transfers from the proton to the nucleus for those events which result in heavy fragment emission. Alpha-particle spectra from events in silver and bromine have been studied, with particular attention to the low energy alpha-particles ( $< 10$  Mev). The abundance of these low energy alpha-particles, which increases sharply with increasing bombarding energy, has long been puzzling in terms of evaporation theory. It has now been established that in nearly every case these alpha-particles are emitted in the backward direction

Table 1

Sensitivity of Mass Spectrometer and Contamination Level of Chemical Purification Procedure for the Detection of Helium and Argon in Metals

Rare gas	Sensitivity of mass spectrometer,* atoms	Contamination level at same mass number, atoms
$\text{He}^3$	$2 \times 10^3$	$3 \times 10^9$
$\text{He}^4$	$2 \times 10^5$	$4 \times 10^{10}$
$\text{A}^{36}$	$1 \times 10^4$	$3 \times 10^{10}$
$\text{A}^{38}$	$1 \times 10^4$	$6 \times 10^9$
$\text{A}^{40}$	$1 \times 10^4$	$8 \times 10^{12}$

\*The sensitivity is the limit of detectability, i.e., the number of atoms required to give a signal equal to the noise level.

with respect to the recoil nucleus. Thus their energies in the center-of-mass system are certainly higher than those measured in the laboratory system. However, corrections for this effect cannot be estimated quantitatively at present because of insufficient knowledge of the momenta of the nuclei at the time of emission.

The frequency of events of type 3 (two fragments with  $Z > 6$ ) increases fairly rapidly with increasing proton energy and reaches about one-fourth of the total number of events at 3 Bev. At least some of these events may be considered fissions, and the angular correlation between the two heavy fragments is being studied. The most probable angle between the fragments appears to be  $\approx 130^\circ$ .

Applications of the recently completed Monte Carlo calculations of nuclear cascade processes to the interpretation of various high energy nuclear reactions form a continuing part of the nuclear chemistry program. In one such study, the results of these calculations have been combined with evaporation calculations to give further insight into the competition between fission and evaporation processes in highly excited heavy nuclei, and into the charge distribution in the primary fission act. Calculations of the average neutron-to-proton ratios of fission products were made with various assumptions about the following parameters: the energy dependence of the relative widths for fission and neutron evaporation, the critical value of  $Z^2/A$  at which fission becomes the dominant process, and the charge distribution between the fission fragments. These calculated results were compared with experimental data on average neutron-to-proton ratios of fission products resulting from the interaction of 8 and 87-Mev protons with  $\text{Th}^{232}$ , and of 450-Mev protons with  $\text{U}^{238}$ . The comparisons show that the so-called equal charge displacement process, hitherto thought to be characteristic

of low energy fission, persists in uranium and thorium at these energies; in this type of fission, the most probable nuclear charges of complementary fragments are equally displaced from the charge values corresponding to stability. Another conclusion from the analysis is that fission in the heaviest nuclei occurs before complete de-excitation by particle evaporation. In fact, the data indicate that fission begins to compete effectively with particle emission when the  $Z^2/A$  value is  $\approx 36$  or greater, in a fashion independent of excitation energy.

The application of high sensitivity mass spectrometry to the investigation of high energy nuclear reaction cross sections has continued. This is important for two reasons: mass spectrometrically determined cross sections for formation of long-lived and stable products can supplement radiochemically measured cross sections, which allows more complete exploration of the systematics of reaction yields; and the yields of certain stable species among the lightest elements, especially  $\text{He}^3$  and  $\text{He}^4$ , can be used to fix some of the parameters in nuclear evaporation theory.

High sensitivity mass spectrometry has been pursued with both surface ionization and electron bombardment sources. The former technique has been extended to a study of cesium yields resulting from the interaction of uranium with protons between 200-Mev and 3-Bev kinetic energy. After irradiation, the bulk of the uranium was removed by ether extraction from a nitrate solution, and the aqueous phase was directly evaporated on the surface ionization filament. By this very simple procedure the contamination with stable cesium was kept to a minimum. In addition to the independent yields of  $\text{Cs}^{129}$ ,  $\text{Cs}^{131}$ ,  $\text{Cs}^{132}$ ,  $\text{Cs}^{134}$ ,  $\text{Cs}^{135}$ , and  $\text{Cs}^{136}$ , chain yields of  $\text{Cs}^{127}$ ,  $\text{Cs}^{129}$ ,  $\text{Cs}^{131}$ ,  $\text{Cs}^{135}$ , and  $\text{Cs}^{137}$  could be determined by analysis of different portions of target material at various time intervals after irradiation. At present, only yield ratios of the various cesium isotopes are known for each bombarding energy, but radiochemical determination of the cross section for formation of  $\text{Cs}^{136}$  (13-day half-life) will permit conversion of these relative yields to absolute cross sections.

The rare gases furnish a number of stable products which can be studied by a high sensitivity mass spectrometer. In measuring the production cross section for stable nuclides, contamination must be kept below  $10^8$  to  $10^{12}$  atoms. This contamination results from the natural elements in

Table 2

Comparison of Helium and Tritium Production in Iron by High Energy Proton Bombardment

Energy, Bev	Cross section, mb		
	$\text{He}^4$	$\text{He}^3$	T
0.16	120	11	7
0.43	450	45	32
3.0	1300	240	100

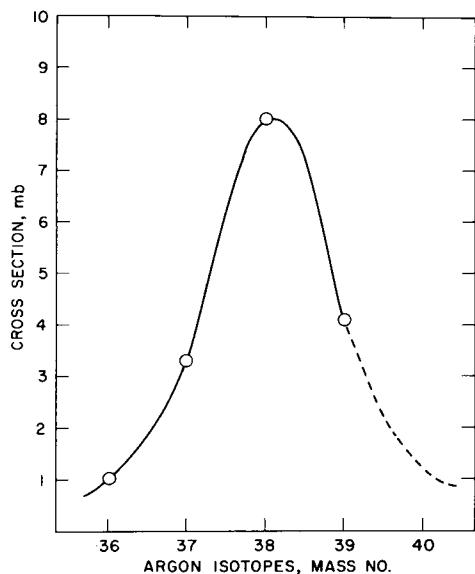


Figure 1. Cross section for the production of argon isotopes in an iron target by 0.43-Bev protons.

the target material and the chemical processing employed. The mass spectrometer and sample processing line have the sensitivities and contamination levels listed in Table 1. This equipment is adequate for the study of  $\text{He}^3$  and  $\text{He}^4$  at currently available synchrocyclotron beam intensities and for the study of argon isotopes when about  $10^{16}$  protons are available.

The cross sections for  $\text{He}^3$  and  $\text{He}^4$  produced in iron by 0.16, 0.43, and 3.0-Bev protons were measured. The results are compared in Table 2 with the tritium cross sections previously measured by Fireman and Schwarzer. An unexpected finding is that the cross section for  $\text{He}^3$  production in iron by proton bombardment is higher than the tritium production cross section in the range 0.16 to 3 Bev.

Current evaporation and cascade calculations do not account for this experimental result. The work at present is being extended to higher and lower values of  $Z$  in order to get a clearer understanding of how the calculations should be modified.

The results for argon production shown in Figure 1 are the first direct experimental results for the yields of stable products for a given  $Z$ . The values fall on a Gaussian curve with a maximum near mass 38.

### Nuclear Processes in Meteorites

Rare gases are produced in iron meteorites by the action of cosmic rays. For this reason the relative cross sections for production of the rare gases from iron targets are related to the relative rare gas contents of iron meteorites. A joint study of the rare gas contents of iron meteorites and the cross sections for production in iron targets bombarded by protons should result in information concerning the constancy of cosmic radiation in time and space, the original size of meteorites before entering the earth's atmosphere, and the length of time meteorites have been in space. Data on such matters should eventually have a bearing on the origin of meteorites and possibly on the origin of the solar system itself.

The cross sections measured bear directly on the rare gases produced by cosmic rays in iron meteorites. From the cross section of directly produced  $\text{He}^3$  relative to the cross section for  $\text{T}$ , previous measurements of the  $\text{He}^3$ - $\text{T}$  exposure ages of meteorites must be reduced by a factor of about 3. From the argon isotope cross sections it is seen that 80% of the  $\text{A}^{36}$  in meteorites is the result of beta decay of  $\text{Cl}^{36}$  produced by cosmic rays; hence, the  $\text{A}^{36}/\text{Cl}^{36}$  ratio should be a reliable method for measuring exposure ages of meteorites.

Table 3

#### Rare Gases in Iron Meteorites

Meteorite	$\text{He}^4$ , (scc/g) $\times 10^6$	$\text{He}^4/\text{He}^3$	$\text{He}^4/\text{A}^{38}$	$\text{He}^4/\text{Ne}^{21}$	$\bar{E}$ , Bev	$\text{He}^4$ production rate, (atoms/year) $\times 10^{-6}$	Exposure age, years $\times 10^{-6}$
Arispe	9.3	2.9	100	460	$>3$	1.5	170
Casas Grandes	3.5	2.9	94	360	$>3$	1.5	63
Toluca	3.0	2.9	81	500	$>3$	1.5	56
Odessa	7.0	3.2	62	300	$\approx 3$	1.3	150
Sikhote-Alin	4.9	3.7	110	330	$\approx 3$	1.3	100
Williamstown	19	4.0	56	320	$\approx 1$	1.0	510
Canyon Diablo	0.06	7.1	50	640	$\approx 0.5$	0.45	3.6

Seven iron meteorites with different amounts of helium were selected. Neon, argon, and helium were then measured in all the samples, with the results shown in Table 3. The average cosmic-ray energy ( $\bar{E}$ ) at the site of the sample was estimated by comparison of the  $\text{He}^4/\text{He}^3$ ,  $\text{He}^4/\text{A}^{38}$ , and  $\text{He}^4/\text{Ne}^{21}$  ratios with ratios obtained from proton bombardment experiments. The ratio involving  $\text{Ne}^{21}$  was estimated from the relative behavior of  $\text{Na}^{24}$ . Fireman and Schwarzer have previously estimated the rate of tritium production at the surface of meteoroids. If it is assumed that the  $\text{He}^4$  production rate is 9 times that for tritium (as suggested by an extrapolation of the values in Table 2 to energies of 6 to 10 Bev), and that it decreases with energy as the cross section does, the values given in Table 3 are obtained for the  $\text{He}^4$  production rate. The cosmic-ray exposure ages listed are obtained by dividing the total amount of  $\text{He}^4$  by the estimated production rate. These estimates, while crude as present, should be correct to at least a factor of 2 or 3, and the relative changes are probably much more accurate.

It is evident that the exposure ages vary greatly - from only a few million years to more than half a billion years. The meteoroids, then, have not all had their present size for the same length of time. This suggests that the asteroids, or other sources of meteoroids, are continually breaking into smaller pieces.

#### Potassium-Argon Ages of Iron Meteorites

Ages of stone meteorites have been determined by several methods which involve the determination of K/A ratios, lead isotope ratios, Rb/Sr ratios, or U/He ratios. These different methods lead to a maximum age of  $5 \times 10^9$  years for stone meteorites. The concentration of elements which may be used for age determination is much smaller in iron meteorites than in stone meteorites. However, neutron activation analysis is such a sensitive analytical tool that it is possible to determine the potassium and argon contents of such meteorites.

For the determination of  $\text{A}^{40}$ , the reaction  $\text{A}^{40}(n,\gamma)\text{A}^{41}$  was used, while potassium was determined by use of the  $\text{K}^{41}(n,\gamma)\text{K}^{42}$  reaction. Meteorite samples were irradiated in the Brookhaven reactor, and the potassium and argon were subsequently obtained by fusion of the samples *in vacuo*. The amounts of potassium and argon were measured by radio-assay techniques, which included

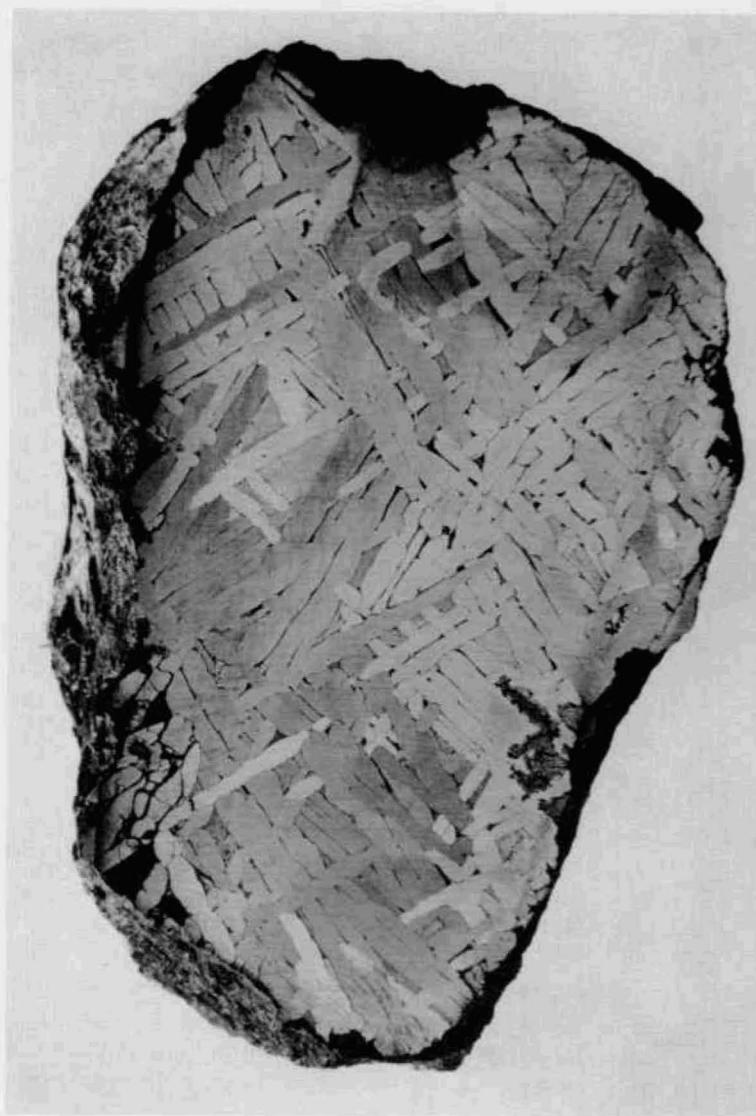


Figure 2. Cross section of a piece from the Toluca meteorite, used in the determination of cosmic-ray ages of meteorites. The sample has been etched to display the Widmanstätten patterns.

comparisons with known standards submitted to the same irradiation. A small correction was made for the  $\text{A}^{40}$  produced by cosmic-ray spallation reactions in the meteorite.

A very large variation was found in the concentrations of potassium and argon in different samples of the same meteorite. However, the ratio  $\text{A}^{40}/\text{K}$  was somewhat more constant, with at most a sixfold variation. The ages of these iron meteorites (or more correctly, the times elapsed since their solidification) were found to range from 5.3

to 13 billion years. Because of the possibility of argon diffusion out of the meteorite, the higher values are considered to be the more significant. This means that the time elapsed since solidification is roughly twice as great for iron as for stone meteorites, which would be consistent with the theory that the parent body of meteorites was composed of a silicate phase and an iron phase. Since most of the radioactivity would be concentrated in the silicate phase, it would have a thermal history different from that of the iron phase.

### Studies of Radionuclides

Experimental and theoretical investigations of the relative probabilities of *K*-capture and positron emission for first-forbidden transitions have been under way at this laboratory for some time in order to derive information about the fundamental beta-decay interactions. A comparison of theoretical and experimental  $K/\beta^+$  ratios for first-forbidden transitions with spin change zero in  $\text{As}^{74}$  and  $\text{Rb}^{84}$  shows the ratios to be greater than that expected for allowed transitions. However, from existing data of several workers the experimental ratio for  $\text{I}^{126}$  appeared to be less than that expected for an allowed transition, in disagreement with theoretical expectations. Since these results were judged to be less reliable than those for  $\text{As}^{74}$  and  $\text{Rb}^{84}$ , a redetermination of the  $K/\beta^+$  ratio for  $\text{I}^{126}$  was made. By use of a triple coincidence method, a direct measurement was made of the fraction of the transitions to the first excited state that occur by positron emission. This eliminated many of the sources of error inherent in the earlier measurements. In addition, since there was some disagreement between various workers as to the positron end-point energies and the level of the first excited state, the positron spectrum was redetermined by using a high-resolution, intermediate-image beta spectrometer. The end-point energy of the ground state transition was found to be  $1.129 \pm 0.005$  Mev. The level of the first excited state was determined from gamma spectra to be  $670 \pm 5$  kev, in good agreement with the result obtained from Coulomb excitation. The  $K/\beta^+$  ratio was found to be  $143 \pm 7$ , whereas the value previously reported was 95. The new value is consistent with theoretical expectations based on the  $K/\beta^+$  ratios obtained for similar transitions.

The discovery of the optical spectrum of technetium in certain types of stars renewed interest several years ago in the problem of the occurrence

of technetium in nature. The most recent geochemical investigations do not support earlier reports of terrestrial technetium. The known half-lives of  $\text{Tc}^{98}$  ( $1.5 \times 10^6$  years) and  $\text{Tc}^{99}$  ( $2.1 \times 10^5$  years) are too short to allow survival of primordial technetium in the earth's crust or stars. However, if the ground state of  $\text{Tc}^{97}$  were very long-lived this would not be the case, and it should be possible to find primordial technetium in nature. Accordingly, a study of the decay properties of  $\text{Ru}^{97}$  and  $\text{Tc}^{97}$  was undertaken. From the analysis of decay curves, a half-life of  $2.88 \pm 0.04$  days was obtained for  $\text{Ru}^{97}$ . By comparison of the *K* x-ray intensity of 90-day  $\text{Tc}^{97m}$  with the *K* x-ray intensity of the  $\text{Ru}^{97}$  parent from which it grew, it was found that  $\text{Ru}^{97}$  branches  $0.041 \pm 0.005\%$  to  $\text{Tc}^{97m}$  and 99.96% to  $\text{Tc}^{97}$ . The half-life of the ground state of  $\text{Tc}^{97}$  was found to be  $(2.6 \pm 0.4) \times 10^6$  years by comparison of its *K* x-ray intensity with that from the 90-day  $\text{Tc}^{97m}$  isomeric state. The *L/K*-capture ratio of  $\text{Tc}^{97}$  appears to be somewhat larger than that for  $\text{Tc}^{96}$ . No gamma-rays attributable to  $\text{Tc}^{97}$  were observed. In view of this experimentally determined half-life of  $\text{Tc}^{97}$ , no primordial technetium can have survived, and that observed in stars must have been formed within the last few million years.

An outgrowth of excitation function studies with the 60-in. cyclotron is the discovery of a new isotope,  $\text{Ge}^{65}$ . This nuclide was produced by the  $\text{Zn}^{64}(\alpha, 3n)\text{Ge}^{65}$  reaction and has been identified by chemical separation of germanium, by milking experiments which establish that it is the parent of 15-min  $\text{Ga}^{65}$ , by excitation function studies, and by cross bombardments.  $\text{Ge}^{65}$  decays with a  $1.5 \pm 0.2$ -min half-life predominantly to the ground state of 15-min  $\text{Ga}^{65}$  by emitting  $3.7 \pm 0.5$  Mev positrons. Gamma-rays having energies of 0.67 and 1.72 Mev are present to the extent of  $\approx 3\%$  and  $\approx 2\%$  of the positrons.

A previously reported neutron-deficient gadolinium isotope having a half-life  $> 20$  days has now been assigned to  $\text{Gd}^{146}$  on the basis of excitation function studies. An activity having a half-life of 45 days was observed in gadolinium chemically separated from targets of  $\text{Sm}^{144}$  which had been bombarded with alpha-particles. The threshold for the production of this activity was  $\approx 23$  Mev, in agreement with that predicted for the  $\text{Sm}^{144}(\alpha, 2n)\text{Gd}^{146}$  reaction. Repeated separation of europium from the  $\text{Gd}^{146}$  yielded a daughter activity with a 4.4-day half-life. The most promi-

inent radiations of  $Gd^{146}$  are  $K$  x-rays, 115-kev gamma-rays, and 150-kev gamma-rays. The gamma spectrum of  $Eu^{146}$  showed  $K$  x-rays and lines at 640 and 740 kev. The last two radiations are in coincidence with each other.

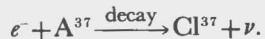
$Ba^{129}$  has been observed as a product of several reactions in the Cosmotron, and, since its decay scheme was not well known, a study of this nuclide was undertaken.  $Ba^{129}$  was prepared by the  $(n,2n)$  reaction on enriched  $Ba^{130}$  targets. Its half-life was measured to be  $2.45 \pm 0.05$  hr, significantly longer than the previously reported value of 1.9 hr. Gamma-rays having energies of 0.127, 0.182, 0.210, and 1.45 Mev were observed, with relative intensities of 33, 100, 57, and 42, respectively. Coincidences were observed only between positrons and the lines at 0.127 and 0.210 Mev, and between the 0.182 and 1.45-Mev radiations. These data, however, are not sufficient to establish a unique decay scheme for  $Ba^{129}$ .

#### Neutrino Detection

An experiment has been in progress at the Savannah River plant during the last two years to determine whether the antineutrinos ( $\bar{\nu}$ ) emitted by an operating nuclear reactor are capable of inverting an electron capture reaction. The negative beta decay of fission products in a reactor produces a high flux of antineutrinos in the vicinity of the reactor. A large volume of carbon tetrachloride was irradiated by this high flux of antineutrinos and the reaction



was looked for by isolation of the  $A^{37}$  and observation of its radioactive decay (34-day half-life) back to  $Cl^{37}$  by electron capture,



In the electron capture decay of  $A^{37}$  neutrinos ( $\nu$ ) are emitted; the experiment tests whether this reaction can be inverted by the antineutrinos ( $\bar{\nu}$ ) from fission products.

The currently accepted two-component theory of the neutrino describes the neutrino and the antineutrino as particles which differ in the orientation of the spin axis with respect to momentum of the particle. This theory would predict a zero rate for the reaction sought in this experiment, since the antineutrinos emitted by the reactor would have a spin orientation opposite to that required to invert the electron capture decay. The

experiment, if carried out with high sensitivity, constitutes a severe test of the two-component theory of the neutrino. More generally, the experiment tests the principle of lepton conservation (leptons are light particles such as electrons, neutrinos, and  $\mu$ -mesons). If the reaction sought for occurred, an electron and an antineutrino would be emitted in a beta decay in the reactor; then the antineutrino would be absorbed in the capture reaction to produce an electron. The over-all reaction would result in the emission of two electrons by this two-step process, in conflict with the principle of lepton conservation.

Experiments have been performed with 1000-gal quantities of  $CCl_4$  contained in two 500-gal tanks. The argon activity was swept from the liquid with helium, separated from the helium gas stream, purified, and placed in a low level Geiger-Müller counter for measurement. Very small counters (0.4 cc) were used to reduce the background (7 to 8 counts/day).

$A^{37}$  has been observed in these experiments, and its identity was confirmed by half-life measurements. The cross section for the capture reaction is

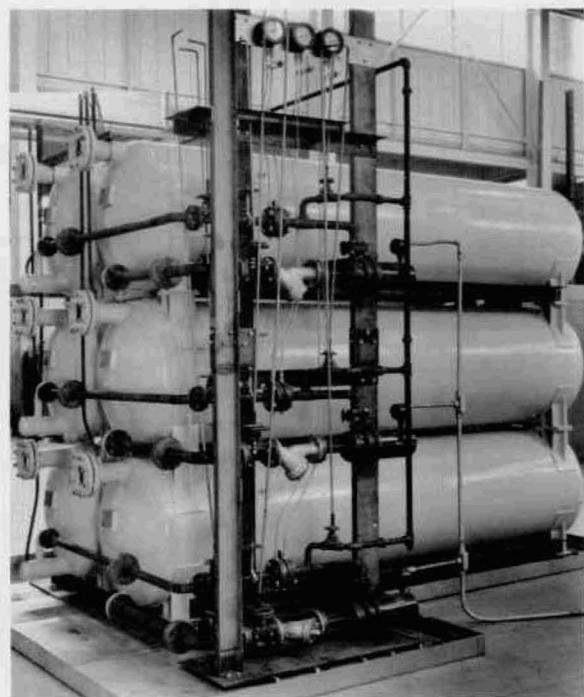


Figure 3. Neutrino detection equipment. The apparatus shown consists of six 500-gal tanks of  $CCl_4$  and circulatory pumps. In the experiment the tanks are immersed in water which serves as a fast neutron shield.

$(1.1 \pm 0.2) \times 10^{-45}$  cm $^2$ , compared to a theoretical cross section of  $5.2 \times 10^{-45}$  cm $^2$ . A $^{37}$  can also be produced in the CCl $_4$  by cosmic radiation and fast neutrons from the reactor. It has not been possible to perform experiments with the reactor off to determine how much of the A $^{37}$  observed was produced by cosmic rays. However, estimates made from cosmic-ray data indicate that part of the observed activity was produced by cosmic-ray  $\mu$ -mesons. Therefore, it is tentatively concluded that the cross section for the Cl $^{37}(\bar{\nu}, e^-)A^{37}$  reaction with fission product antineutrinos is less than or equal to  $1.1 \times 10^{-45}$  cm $^2$ .

During the past year it was considered important to extend these experiments to higher sensitivity and to test background effects from cosmic radiation and fast neutrons. A new apparatus (Figure 3) which contains 3000 gal CCl $_4$  was built,

tested, and finally installed at the Savannah River plant. The apparatus was designed to hold the maximum amount of CCl $_4$  that could safely be placed at a location of high antineutrino flux in the Savannah River plant. The apparatus consists of six 500-gal cylindrical tanks 34 in. in diameter and 12 ft long, arranged horizontally in a stack of three pairs of tanks. Each pair of tanks is stirred separately by a 50-gal/min pump. The entire assembly of tanks is placed in a large steel tray that can be filled with water to serve as a fast neutron shield. Measurements were started with the new apparatus in the middle of April and are still in progress.

#### High Precision Mass Measurements

The mass synchrometer, a high resolution mass spectrometer developed in the Physics Depart-

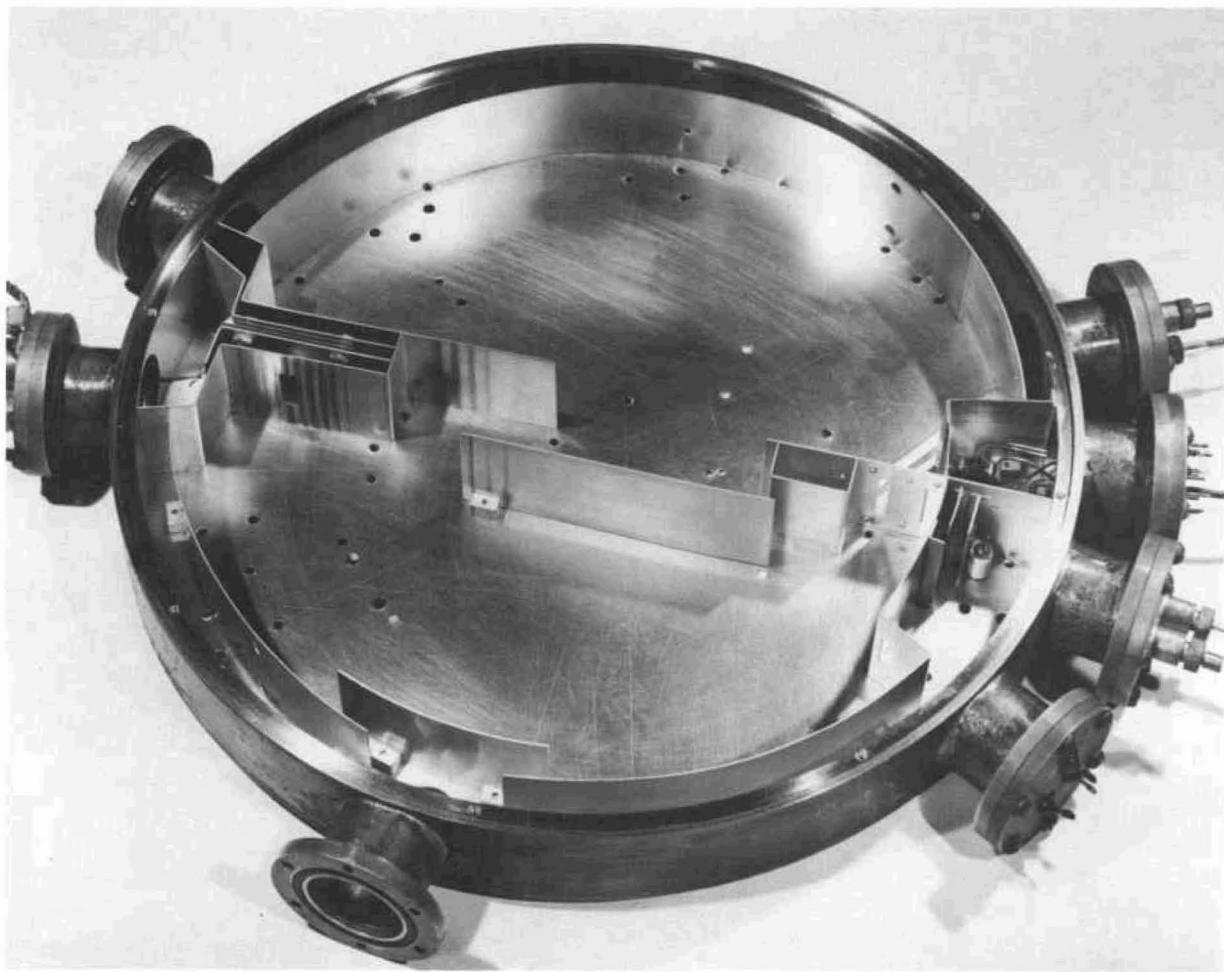


Figure 4. Vacuum chamber of the mass synchrometer.

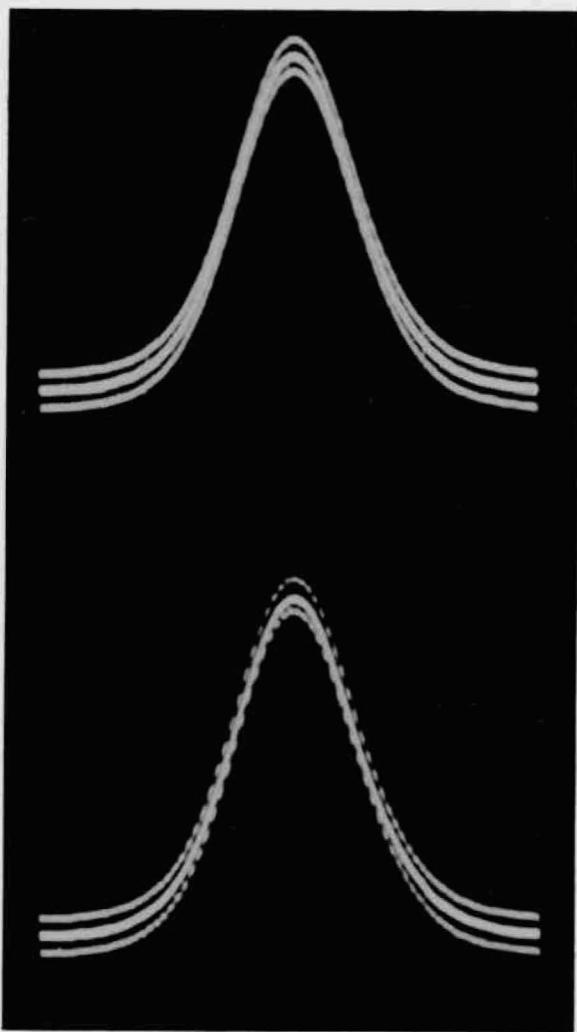


Figure 5. Oscilloscope presentation of mass synchrometer output, showing matched peaks of the  $\text{He}^3$ -HD doublet.

ment, is now being used by the Chemistry Department to measure precise mass differences in beta decay. These measurements, when combined with experimentally determined beta end points, may be used to establish experimental upper limits for the neutrino rest mass. Furthermore, within the limits of uncertainty in the neutrino rest mass, a sensitive comparison of mass differences obtained by widely differing techniques is made possible.

The  $\text{T-He}^3$  mass difference was determined by measuring the mass differences of the three doublets,  $\text{HD-He}^3$ ,  $\text{D}_2\text{-HT}$ , and  $\text{C}_2\text{H}_4\text{-C}_2\text{D}_2$ . The mass difference determined was  $18.65 \pm 0.20$  kev, where the quoted error is a standard error. If the value of  $18.1 \pm 0.2$  kev for the tritium beta end point is subtracted from this mass difference, a neutrino

rest mass of  $0.55 \pm 0.28$  kev is obtained. It should be noted that the value used for the beta end point is a weighted average of extrapolated values ranging from 17.6 to 18.9 kev and that the extrapolated end point includes the neutrino rest mass. In view of the experimental error indicated by the spread of the reported data, this procedure (i.e., setting extrapolated values equal to true beta end points) was considered valid in estimating the magnitude of the neutrino rest mass. The data establish an upper limit on the rest mass of the order of 1 kev. The lower limit depends very strongly on the accuracy of the synchrometer data and the beta end-point measurements.

By the combination of measured values of the three doublets with the value  $M_{\text{H}} - 1 = 8145.39 \pm 0.11 \mu\text{MU}$ , mass defects for  $M_{\text{T}} - 3$  and  $M_{\text{He}^3} - 3$  are obtained, which are in agreement with values obtained from nuclear reaction data but considerably more precise.

Measurements on the  $\text{C}^{14}\text{-N}^{14}$  mass difference are under way. Preliminary data indicate a mass difference in agreement with the 155-kev beta end point and consistent with the 1-kev upper limit for the neutrino rest mass.

#### CHEMICAL EFFECTS OF NUCLEAR TRANSFORMATIONS IN INORGANIC SYSTEMS

Investigations of the behavior of recoil atoms in solids have been continued. Several systems have been examined, including arsenic compounds, *cis*- and *trans*-dichloro-bis(ethylenediamine) cobalt (III) nitrate, and ferricinium picrate.

When arsenic pentoxide was irradiated with neutrons,  $\approx 41\%$  of the radioactive arsenic was found as  $\text{As(V)}$ , while only 4% was found in this oxidation state when arsenic trioxide was irradiated. In each case the amount of activity in the parent form was increased by heating the irradiated samples.

When irradiated samples of *cis*- or *trans*-dichloro-bis(ethylenediamine) cobalt(III) nitrate were dissolved in water,  $\approx 70\%$  of the radioactive cobalt was separable as  $\text{Co(II)}$ . Also for both isomers  $\approx 20\%$  of the radioactive cobalt was retained in the parent form, and  $\approx 1\%$  was found as the other isomer. These results indicate that the radioactive cobalt tends to re-form the parent species rather than other possible configurational isomers.

In samples of irradiated ferricinium picrate,  $\approx 2\%$  of the radioactive iron was found in the form

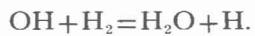
of the parent compound, and  $\approx 69\%$  as inorganic iron. In the case of irradiated ferrocene,  $\approx 12\%$  of the radioactive iron was found as ferrocene. In contrast to ferrocene, ferricinium picrate does not undergo thermal annealing.

A model based on the "displacement spike" concept has met with some success in explaining recent results of Szilard-Chalmers investigations. Various models proposed to explain the results of thermal annealing experiments have also been considered. These models treat the recombination of fragments having small initial separations, as well as the recombination of randomly distributed fragments. In this respect, the change in thermal annealing properties produced by radiation damage in a solid presents an interesting problem.

### CHEMICAL EFFECTS OF RADIATION

#### Aqueous Systems

Chemical changes in irradiated aqueous solutions result from the decomposition of water into oxidizing and reducing free radicals which have generally been called OH and H. These are formed in regions of high concentration along the track of the ionizing particle, and a fraction of them combine with one another to form  $H_2$  and  $H_2O_2$ , the water decomposition products, while the remainder diffuse out into the solution and make their presence known by reacting with dissolved materials. However, work on solutions containing dissolved oxygen, hydrogen peroxide, and hydrogen has shown that it is necessary to be somewhat sophisticated in considering the real character of these radicals. The OH radicals clearly are capable of oxidizing hydrogen molecules, as shown by numerous experiments on various systems, and the product of this reaction is certainly expected to be a hydrogen atom:



Experiments in which solutions containing both oxygen and hydrogen peroxide as well as hydrogen are irradiated with  $\gamma$ -rays show that the H atoms arising from this reaction act preferentially on the oxygen and reduce it to hydrogen peroxide, and the hydrogen peroxide itself is not attacked by these H atoms until all the oxygen present has been consumed. However, in solutions containing only oxygen and hydrogen peroxide with little or no hydrogen present, both oxygen and hydrogen

peroxide are reduced under gamma irradiation. The reducing radicals from water must act on both these compounds, and the reactions have naturally been attributed to H atoms from water decomposition:



and



The conclusion seems inevitable that the "H atom" arising from decomposition of water under irradiation is different from the "H atom" formed by free radical oxidation of  $H_2$ . The possibility of different forms may arise from the polarity of water which allows solutes to exist in acidic or basic forms corresponding to the loss or gain of a proton. For the H atom the acidic and basic forms would be, respectively, the ion  $H_2^+$  and the solvated electron:



The relative stability of hydrogen peroxide in acid solutions under irradiation suggests that the form resulting from oxidation of  $H_2$  may be the form more stable in acid solutions. Possibly the initial product of oxidation of  $H_2$  by a free radical is not atomic H but the ion  $H_2^+$  resulting from transfer of an electron from the  $H_2$ ; the initial radiolysis product of water could then be supposed to be a true H atom. Alternatively, the so-called H resulting from water radiolysis could be a solvated electron, while the product of oxidation of  $H_2$  would be a true H atom.

Recognition of the possibility that H atoms may exist in two forms with different relative reaction rates towards  $O_2$  and  $H_2O_2$  has clarified a considerable mass of previously published data on decomposition of water by various types of radiation. For example, oxygen, hydrogen peroxide, and hydrogen all appear when water is exposed to densely ionizing radiations, and the amount of decomposition proceeds linearly with time even when the products are allowed to accumulate in the water. This has been shown to result naturally from the competitive reactions of the free radicals with the products. The relative amounts of the three products formed with different kinds of radiation have been predicted quantitatively from the known characteristic radical and molecular yields and from the relative rates of reaction of radicals with the different products. These rates were obtained

from studies of solutions of oxygen, hydrogen, and hydrogen peroxide under gamma irradiation. If the irradiated water initially contains oxygen or hydrogen peroxide, the linearity of growth of the products with time is completely destroyed, and complicated kinetics are found which have also been explained quantitatively. Some early studies of effects of  $\gamma$ -rays and of reactor radiation on solutions containing hydrogen peroxide and hydrogen showed that when the hydrogen was in excess the net reaction was simply



and that the higher the concentration of hydrogen peroxide, the more slowly this reaction proceeded. If the initial hydrogen peroxide concentration was increased beyond that of the hydrogen, however, the rate passed through a minimum and increased at higher hydrogen peroxide levels, while appreciable quantities of oxygen appeared as a product. These observations have now been quantitatively explained by use of the concept of two kinds of "H atoms" which give the same chemical reactions, but react at different relative rates with oxygen and hydrogen peroxide.

Work has continued on the gamma irradiation of ferrous chloride solutions and potassium nitrite solutions, and the kinetic complexities of these systems have been largely unraveled and clarified in terms of free radical reactions. The yield of molecular hydrogen is affected by the concentration of oxidizing agents in a similar fashion in neutral and in highly acid solutions, even though the character of the H atoms present may be different in the two cases. Some apparent anomalies in the characteristics of nitrite oxidation are ascribed to participation of the reaction product, nitrate ion, in the reaction with H atoms, and the whole reaction system that results from irradiation of nitrite or nitrate solutions is gradually becoming clearer.

#### Hydrocarbons Adsorbed on Solids

Continuation of the study of radiolysis of pentane adsorbed on silica gel emphasizes the differences in the effect of radiation on this compound in the adsorbed state as compared to that on the ordinary bulk material. Analysis of the hydrocarbon products by gas chromatography shows that the formation of unsaturated compounds is completely suppressed when the pentane is adsorbed on silica gel, but large yields of branched chain compounds were formed that do not occur in the

ordinary bulk irradiation. The total quantity of pentane decomposed by a given dose of gamma radiation is many times greater in the adsorbed than in the bulk state, which probably means that energy originally given up to the solid by the radiation can be transferred to the organic molecules and produce decomposition. The work has now turned to a study of the irradiation of pentane adsorbed on compounds of the "molecular sieve" type. Results appear to show a pronounced increase in the yield of hydrogen in the adsorbed state, but no effect on the yield of methane, which indicates that the solid may strongly affect certain modes of radiolytic decomposition but leave other modes unaffected.

#### CHEMISTRY OF ISOTOPES

The differences in the thermodynamic properties of isotopes, as well as in their rates of chemical reaction, can be correlated with the effect of isotopic substitution on the molecular vibrations. General theorems concerning the effect of isotopic substitution on the vibrational frequencies are of interest in understanding the molecular vibrations, the forces between the atoms in molecules, and the differences in chemical properties of isotopes. During the past year, a new theorem has been found concerning the vibrational frequencies for a homologous series of isotopic molecules, including equivalent isotopic isomers. This theorem, the second-order sum rule, facilitates the analysis of the molecular spectra of polyatomic molecules. It also extends the limits of justification of the rule of the geometric mean (e.g.,  $Q_{\text{HDO}}^2 = Q_{\text{H}_2\text{O}}Q_{\text{D}_2\text{O}}$ , where  $Q$  is the partition function).

Through the use of the product rule for the molecular vibrations belonging to different symmetry classes, it is possible to simplify the treatment of the differences in chemical properties of linear isotopic molecules.

#### The Second-Order Sum Rule

Several years ago the sum rule for vibrational frequencies of isotopic molecules was formulated by Decius and Wilson, and independently by Sverdlov. An example of this rule is

$$2\sum\lambda_i(\text{HDO}) = \sum\lambda_i(\text{H}_2\text{O}) + \sum\lambda_i(\text{D}_2\text{O}),$$

where  $\lambda$  is proportional to the square of the vibrational frequency. An extension of this type of relation has been found: In a homologous series of

isotopic molecules (e.g.,  $\text{CH}_4$ ,  $\text{CH}_3\text{D}$ ,  $\text{CH}_2\text{D}_2$ ,  $\text{CHD}_3$ , and  $\text{CD}_4$ ), the differences in  $\sum \lambda_i^2$  depend only on the interactions between atoms not bonded by the usual valence bonds. If the molecules are so chosen that there is an equivalent number of such interactions, then such a set of molecules will have identical sums of  $\sum \lambda_i^2$  and will obey the rule of the mean through powers of  $(\hbar/kT)^4$ . Some examples which have been given in detail are:

$$\begin{aligned} & 8\sum \lambda_i^2(\text{H}_2\text{C}=\text{CHD}) + \sum \lambda_i^2(\text{D}_2\text{C}=\text{CD}_2) \\ & = 3\sum \lambda_i^2(\text{H}_2\text{C}=\text{CH}_2) + 2\sum \lambda_i^2(\text{H}_2\text{C}=\text{CD}_2) + \\ & \quad 2\sum \lambda_i^2(\text{cis-HDC}=\text{CDH}) + \\ & \quad 2\sum \lambda_i^2(\text{trans-HDC}=\text{CDH}) . \end{aligned} \quad (1)$$

$$\begin{aligned} & \sum \lambda_i^2(1,2,4\text{-C}_6\text{H}_3\text{D}_3) + \sum \lambda_i^2(o\text{-C}_6\text{H}_4\text{D}_2) \\ & = \sum \lambda_i^2(1,2,3\text{-C}_6\text{H}_3\text{D}_3) + \sum \lambda_i^2(p\text{-C}_6\text{H}_4\text{D}_2) . \end{aligned} \quad (2)$$

$$\begin{aligned} & 2\sum \lambda_i^2(\text{CHD}_3) + \sum \lambda_i^2(\text{CH}_4) \\ & = 2\sum \lambda_i^2(\text{CH}_3\text{D}) + \sum \lambda_i^2(\text{CD}_4) . \end{aligned} \quad (3)$$

$$\begin{aligned} & 2\sum \lambda_i^2(\text{C}^{13}\text{O}^{16}\text{O}^{18}) + \sum \lambda_i^2\text{C}^{12}\text{O}^{16}_2 + \sum \lambda_i^2\text{C}^{12}\text{O}^{18}_2 \\ & = 2\sum \lambda_i^2(\text{C}^{12}\text{O}^{16}\text{O}^{18}) + \sum \lambda_i^2\text{C}^{13}\text{O}^{16}_2 + \\ & \quad \sum \lambda_i^2\text{C}^{13}\text{O}^{18}_2 . \end{aligned} \quad (4)$$

In each of these relationships, the equality holds for the symmetry classes common to all the molecules in the equation.

### Temperature Independent Isotope Effects in Linear Molecules

In a linear molecule the molecular vibrations can be divided into two classes: stretching vibrations along the internuclear axis, and bending vibrations perpendicular to the axis. The frequencies of the stretching vibrations are usually high, while the bending vibrations, being associated with weak forces, are of low frequency. The bending vibrations are, therefore, nearly classical in their behavior. According to the Teller-Redlich product rule, the product of the vibrational frequencies of one isotopic species is related to the product for a second species by a function of moments of inertia and atomic masses. This rule, combined with the separation of stretching and bending vibrations, leads directly to the result that the temperature independent difference in the

chemical properties of isotopic molecules depends solely on the molecular weights of the molecules. Some consequences of the theorem are: 1) the largest temperature independent effects are to be found with the isotopic  $\text{H}_2$  molecules; 2) the temperature independent effect is not affected by the arrangement of atoms in a linear molecule, i.e., it is the same for  $\text{N}^{15}\text{N}^{14}\text{O}$  and  $\text{N}^{14}\text{N}^{15}\text{O}$ , etc.; 3) the deviations from the rule of the geometric mean for linear molecules are such that symmetrical molecules will always be favored somewhat more than random choice predictions; and 4) the equivalence of certain aspects of the collision and transition state theories of chemical reaction rates is established.

## ORGANIC CHEMISTRY

### Carbon-14 and Carbon-11 Recoil Chemistry

A number of new organic systems have been studied to elucidate the mechanism of action of a recoil fragment in re-entering organic combination after its formation. Information from these studies allows more efficient application of nuclear recoil as a tool for labeling organic molecules.

The effects of change in phase and variation of nitrogen source (in  $\text{C}^{14}$  work) have shown that chemical effects are of primary importance in determining product distribution in recoil systems. Purely physical mechanisms are unable to account

Table 4

#### Ring and Methyl Group Entry of Recoil $\text{C}^{14}$

Product	Radiochemical yield
Toluene (from toluene + 2-methylpyrazine as nitrogen source)	 — $\text{CH}_3$ $\approx 2.2\%^*$ $0.28\%^*$
Aniline (from aniline)	 — $\text{NH}_2$ $\approx 2.8\%$
Toluene (from aniline)	 — $\text{CH}_3$ $\approx 0.01\%$ $\approx 0.06\%$

\*Corrected to 0% nitrogen source.

for product distribution and for the distribution of activity in the isolable compounds.

The irradiation of pure aniline was carried out in order to compare the relative ease of replacement of carbon and nitrogen. A comparison of aniline with toluene (in which toluene plus a nitrogen source is irradiated) should show an equal amount of activity in the methyl groups of the radioactive toluene products from these two sources if they are formed in the hot region by "knock-on" mechanisms. The results of this study are given in Table 4.

It is clear that the replacement of ring carbons is about equally efficient in both toluene and aniline, but that replacement of the methyl carbon in toluene is about five times as efficient as replacement of an amino group in aniline. The masses of these groups are 15 and 16, respectively, and therefore these groups should be comparable in their ability to stop a recoil fragment. The observed difference is a reflection of the strong influence of chemical forces in the product-determining steps of the process.

A study of the formation of benzene-C<sub>11</sub> and of toluene-C<sup>11</sup> formed by the fast neutron irradiation of benzene was carried out in order to investigate the phase effect and the effect of added radical scavengers. The Brookhaven cyclotron was used, the source of fast neutrons being the Li<sup>7</sup>(d,n)Be<sup>8</sup> reaction. Carrier toluene was added and then separated from the benzene by gas-liquid chro-

matography (GLC). The GLC purification method allowed a high degree of purification to be achieved in the short time periods available for work with C<sup>11</sup>. The results of this study are listed in Table 5.

The phase effect is generally used to distinguish between hot and thermal reactions. Hot reactions, caused by recoil fragments with a substantial fraction of the initial recoil energy, should be insensitive to a change in phase or a decrease in temperature. The contribution of thermal reactions should decrease in the solid phase, since these reactions depend on the diffusion and recombination of radical fragments. The data in Table 5 indicate that benzene formation results from both types of reaction, while toluene formation is brought about exclusively by hot reactions. The effect of added radical traps is consistent with this reasoning, since radical traps might not be expected to affect "hot" yields. These results are not explicable by a simple physical impact theory. Consideration of the nature of the reactive fragment, its energy, and its chemical environment should provide a more fruitful approach to product determination in these systems.

Normal pentane was irradiated with thermal neutrons in the presence of several nitrogen sources. Isopentane and all the isomeric hexanes were added as carriers. The results are summarized in Table 6.

The radiochemical yields of synthesis products are comparable to those found in aromatic systems, particularly when considered in terms of replaceable hydrogens. For example, the yield of

Table 5

C<sup>11</sup> Activity Produced by Fast Neutron Irradiation of Benzene

Material irradiated	Radiochemical yield, %	
	Benzene	Toluene
Liquid benzene	4.6	2.3
Solid benzene at	0°	2.5
	— 78°	2.6
	— 195°	3.1
Liquid benzene + DPPH*	2.9	2.3
Liquid benzene + 1,3,5-TNB**	4.1	2.6

\*Diphenylpicrylhydrazine.

\*\*1,3,5-Trinitrobenzene.

Table 6

Radiochemical Yields for the N<sup>14</sup>(n,p)C<sup>14</sup> Reaction in Pentane

Compound	Radiochemical yield, %		
	Run 1*	Run 2**	Run 3†
Isopentane	0.59	—	2.0
Pentane	0.9	1.88	1.56
2,2-Dimethylbutane	<0.01	<0.16	
2,3-Dimethylbutane	<0.01	<0.16	
2-Methylpentane	2.89	2.9	
3-Methylpentane	1.41	1.35	2.25
n-Hexane	3.50	4.38	5.5

\*53 M % n-pentane, 47 M % 2-methylpyrazine.

\*\*80 M % n-pentane, 20 M % 2-methylpyrazine.

†95 M % n-pentane, 5 M % aniline.

3-methylpentane is about 1.5%, while the yield of *m*-xylene from toluene is 1.2%. It has been postulated that the behavior of the reaction fragment is analogous to that of a photolytically produced carbene ( $\text{CH}_2:$ ). This is consistent with the results listed in Table 6. The recoil fragment is indiscriminate in its attack on the molecule, and the yields of synthesis products are in close agreement with those predicted for statistical replacement of hydrogen (see Table 7).

Adequate purification of the alkanes could be brought about only by the use of GLC columns operating in the 1000 to 2000 plate range.

The degradation of toluene formed by the irradiation of benzene plus 2-methylpyrazine showed that  $\approx 88\%$  of the activity was in the methyl group and  $\approx 12\%$  was in the ring carbons. As a first step in testing several mechanisms that might be consistent with this observation, the methyl-labeled toluene was irradiated with 1.9-Mev electrons to determine whether an ionized toluene molecule in the liquid phase could undergo a reversible molecular rearrangement leading to partially ring-labeled toluene. A 17-g sample of toluene absorbed  $\approx 10^{24}$  ev. From the activity level of the sample it would have been possible to detect 0.05% rearrangement, which corresponds to a *G* value for rearrangement of about 0.004 molecules/100 ev. No rearrangement was detected; therefore these values are upper limits on the process in question. While this applies only to such rearrangement as an irradiation by electrons could bring about, it is reasonable to assume that gamma irradiation would lead to the same result. Possible neutron effects remain to be tested. This will allow an experimental distinction to be drawn between a radiation effect and a recoil effect. Similar studies on labeled cycloheptatriene are in progress.

### Reaction Mechanisms

Research in physical organic chemistry during the past year was centered for the most part on two areas: aromatic hydrogen exchange reactions, and acidity measurements in strongly acid solutions. The exchange reactions were used chiefly as a tool in the solution of problems ranging from steric hindrance to secondary kinetic isotope effects, but also proved useful in the acidity determinations. The latter were done largely as an end in themselves, but the techniques used were also applied to the problem of determining the

Table 7  
Ratios for Methyl Substitution Reactions  
in Pentane

Product	Statistical		
	replacement	Run 1	Run 2
3-Methylpentane	1	1	1
2-Methylpentane	2	2.1	2.0
<i>n</i> -Hexane	3	3.2	2.5

acidity dependence of rate-determining proton transfers.

In addition to the two generally recognized mechanisms by which acid-catalyzed reactions may occur in aqueous solution, the *A*-1 and *A*-2 reactions, a third, the *A*-*S<sub>E</sub>*2 reaction, is possible in principle. This reaction is different from the other two in that the proton transfer step rather than some subsequent process is rate determining. The influence of medium on rate is virtually the only means of distinguishing between these important mechanisms. Hence, the question of how changes in the medium, such as those encountered in strongly acid solution, will affect the rate of the *A*-*S<sub>E</sub>*2 reaction is currently of much interest.

Experimental determination of the acidity dependence of the *A*-*S<sub>E</sub>*2 reaction is difficult, because to date no example has been discovered of a rate-determining proton transfer reaction proceeding at a rate sufficiently slow in strong acid. However, a consideration of the keto-enol equilibrium furnishes a solution to this problem. It is well known that the ketone enolization reaction proceeds by a mechanism of the *A*-2 type, and that its rate of reaction is proportional to the concentration of hydronium ion. Simple application of the principle of microscopic reversibility and the Brønsted equation for assessing medium effects on reaction velocities then leads irrevocably to the conclusion that the ketonization of enols is an *A*-*S<sub>E</sub>*2 reaction whose rate is proportional to the hydronium ion concentration. This is confirmed in practice by the fact that the enol content of ketones is independent of acidity over a wide range of acid concentration.

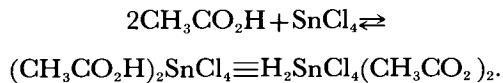
Values of Hammett's acidity function, *H<sub>0</sub>*, for aqueous solutions of strong mineral acids are now fairly well established, but the situation is not the same for weak acids, especially organic acids. This

acidity function was therefore measured in aqueous solutions of dichloroacetic acid, trifluoroacetic acid, and potassium hydrogen sulfate. The aromatic hydrogen exchange reaction, whose mechanism is now well established and whose rate is known to follow  $H_0$ , was then used to demonstrate that in these media under certain conditions the Zucker-Hammett hypothesis is useful as a criterion of mechanism.

This same reaction was used to elucidate the nature of steric interactions between the methoxyl groups of 1,2,3-trimethoxybenzene. The rate of nuclear hydrogen exchange in this material is significantly less than the rate calculated in the usual manner from the partial rate factors for hydrogen exchange in anisole. This suggests that the 1- and 3-methoxyl groups quite effectively shield from reaction the two nuclear positions immediately adjacent. This steric shielding seems to be much more important in decreasing the rate than the loss of configuration favorable for resonance, again for steric reasons, by the 2-methoxyl group.

Secondary hydrogen isotope effects (kinetic isotope effects in reactions not involving rupture of the isotopically substituted bond) have recently been given an explanation involving hyperconjugation. This, however, is inconsistent with another fact uncovered through use of the aromatic hydrogen exchange reaction: This reaction, in which hyperconjugation most certainly plays a major role, exhibits no secondary isotope effect; that is, nuclear hydrogen exchange in toluene and toluene- $\alpha$ - $d_3$  occurs at exactly the same rate.

Acetic acid exhibits enhanced acidity when stannic chloride is added to the pure acid. The question immediately arises whether this increased acidity is caused by the stannic chloride acting as a Brønsted acid or a Lewis acid. The fact that stannic chloride also accelerates the rate of aromatic hydrogen exchange reactions in acetic acid furnishes proof that this is the result of Brønsted acidity. The formation of this new Brønsted acid can be represented by the following equation:



An important but infrequently studied factor affecting ionic reactions in solvents of low dielectric constant is the relative reactivity of free ions

and of ion pairs held together by electrostatic forces. To determine the effect of ion pair formation on the reactivity of bromide ion, a study has been made of the kinetics of the bromine exchange reaction between *p*-nitrobenzyl bromide and potassium bromide or tetraethylammonium bromide in liquid sulfur dioxide. The tracer used in this work was  $\text{Br}^{82}$ . The exchange data have been analyzed in conjunction with known ion pair dissociation constants obtained from conductivity data. In this way, rate constants were obtained for the reaction of *p*-nitrobenzyl bromide with free bromide ion, the  $\text{K}^+\text{Br}^-$  ion pair, and the  $(\text{C}_2\text{H}_5)_4\text{N}^+\text{Br}^-$  ion pair. At 0°C, the same value of the rate constant for free bromide ion was obtained from the data for both potassium bromide and tetraethylammonium bromide. This rate constant is about 15 times as large as that for the  $\text{K}^+\text{Br}^-$  ion pair, and about 4 times as large as that for the  $(\text{C}_2\text{H}_5)_4\text{N}^+\text{Br}^-$  ion pair. The data are consistent with the assumption that the state of dilute solutions of an electrolyte indicated by conductivity data is an accurate representation of its state with respect to chemical reactivity. Rate data have also been obtained for potassium bromide solutions at -10.2° and +10.2°C, so that temperature effects can be investigated.

#### Biosynthesis of Nicotine

When tritium-labeled nicotinic acids are fed to roots of several varieties of tobacco growing in sterile culture media, the activity is found in substantial amounts in the pyridine ring of the nicotine and anabasine produced by the roots. The 2-tritiated acid is incorporated into anabasine, and 2- and 5-tritiated nicotinic acids are found in nicotine (anabasine has so far been examined only with the 2-labeled acid). However, 6-tritiated nicotinic acid is incorporated into nicotine to less than one-tenth the extent of the other two acids. Results from feedings of nicotinic acid-4- $d_1$  have now shown incorporation into the nicotine molecule comparable to that found with the 2- and 5-labeled acids.

Oxidation of the 2- and 6-tritiated acids to the 2- and 6-pyridones by alkaline potassium ferricyanide has shown that substantially all the activity is in the expected position in the two acids. Similar degradation of the nicotine produced by feeding the 2-tritiated acid has shown that the activity is still in the 2-position in the nicotine molecule, and that therefore no exchange or scram-

bling of the activity occurs during the conversion from the acid into nicotine. Formation of a symmetrical intermediate (e.g., pyridine) is also precluded.

These results indicate the likelihood that the incorporation takes place via an oxidative attack on the 6-position of the nicotinic acid molecule, followed by some type of displacement of the carboxyl group at the 3-position by a 5-carbon fragment (apparently derived from ornithine). This postulate is currently being investigated by feedings of  $N^{15}$ -labeled 6-hydroxynicotinic acid, and of several possible nonnitrogenous precursors of this acid. Since the "normal" biological precursors of nicotinic acid are not incorporated into nicotine by the tobacco roots, 6-hydroxynicotinic acid rather than nicotinic acid itself may be the nicotine precursor in the direct synthetic chain, but the tobacco roots may nevertheless be capable of oxidizing nicotinic acid to the actual intermediate compound. Current experiments should shed considerable light on this point.

## STRUCTURAL CHEMISTRY

### Solutions of Proteins at Low Temperatures

At temperatures low enough to prevent the occurrence of irreversible reactions, reversible processes and weakly bonded complexes and intermediates can be studied. For this reason, a search was made for solvents which would dissolve proteins at  $-78^{\circ}\text{C}$  or below and form glasses persisting down to  $-196^{\circ}\text{C}$ . With such solutions in the glassy state, solute properties could be investigated by spectrometric methods. Another requirement was that the solvent be removable at low temperatures, so that the protein could be recovered and assayed in aqueous solution for biological activity.

One such solvent, consisting of 20% methylhydrazine, 40% methylamine, and 40% trimethylamine, was a liquid down to  $-150^{\circ}\text{C}$  and formed a clear glass at lower temperatures. At  $-78^{\circ}\text{C}$ , it could be distilled from a protein solution in a few hours. Another solvent, a mixture of 20% trifluoroacetic acid, 55% methylamine, and 25% trimethylamine, was a viscous liquid down to  $-110^{\circ}\text{C}$ , and a clear glass at lower temperatures. However, it was not removable from the solution at  $-78^{\circ}$ , apparently because of reaction between the acid and the amines.

Proteins studied were acetylated trypsin, catalase, chymotrypsin, chymotrypsinogen, desoxyribonuclease, insulin, lysozyme, ovomucoid, pepsin, peroxidase, ribonuclease, soybean trypsin inhibitor, trypsin, and trypsinogen. These were soluble at  $-78^{\circ}$  to the extent of  $\approx 10^{-4}$  g/ml.

Both fluorescence and phosphorescence spectra of these protein solutions were obtained for mobile solutions at  $-78^{\circ}\text{C}$  and vitreous solutions at  $-196^{\circ}\text{C}$ . With exciting radiation of wavelength 2850 to 3150 Å, spectra were obtained during excitation and with a delay of about 4 msec.

In general, the spectra of the solutions varied with the proteins, and for a given protein varied with solvent. There was no detailed structure in the few spectra taken at  $-78^{\circ}\text{C}$ , and no phosphorescence was visible from these fluid solutions. Since phosphorescence spectra require rigid systems, it would seem that the rigidity within the protein molecule even at  $-78^{\circ}\text{C}$  is not sufficient to permit the trapping of excited states or of electrons. At  $-196^{\circ}\text{C}$ , the glasses exhibited spectra having some structure which was especially marked in the afterglow (delay of 0.004 sec or longer). A correspondence was found between the fluorescence and phosphorescence spectra and the known chemical composition of a protein. Ribonuclease and insulin, which possess no tryptophan residue, did not exhibit phosphorescence in the spectral region characteristic of tryptophan; however, fluorescence and phosphorescence were strong in the region characteristic of the tyrosine residue known to be a constituent of these substances.

The result of Debye and Edwards that aqueous solutions of some proteins exhibited almost indistinguishable phosphorescence spectra was confirmed. However, differences were found in the spectra of some proteins in these solutions, in addition to those ascribable merely to the presence or absence of particular phosphorescent centers.

Instances were encountered in which the protein in the organic medium had a phosphorescence spectrum different from that in aqueous solution, but upon replacement of the organic solvent with water the phosphorescence characteristic of its aqueous solution reappeared unchanged. Such restoration did not occur when the combined amine solvents had been employed. The exclusion of methylamine led, after replacement of the solvent by water, to a restored spectrum deviating but slightly from that of the usual aqueous solutions.

### Partial Molal Volumes and Free Energies of Single Ions

A considerable amount of thermodynamic data is available for electrolytes in aqueous solution, e.g., free energies and partial molal volumes. Recently, various attempts have been made to assign values to single ions and to correlate these values with ionic properties such as the crystal radius ( $r$ ) and charge ( $Z$ ). These attempts have not been entirely satisfactory because implausible physical models and unreliable ionic parameters have been used.

The partial molal volumes and free energies of single ions at infinite dilution have been obtained by the assumption that the ion-solvent interactions are independent of the sign of the charge for large, univalent ions. The partial molal volume of an ion may be expressed as the difference between the intrinsic volume of the ion and the electrostriction of the surrounding solvent. Molal volumes of large, univalent, monatomic ions can be calculated with two assumptions: 1) the ionic radius in solution is larger than the crystal radius by a constant factor; and 2) the electrostriction of the solvent is proportional to  $Z^2/r$ . An almost quantitative account of the volumes of monatomic ions of higher charge can be given, if it is assumed that the electrostriction becomes independent of size.

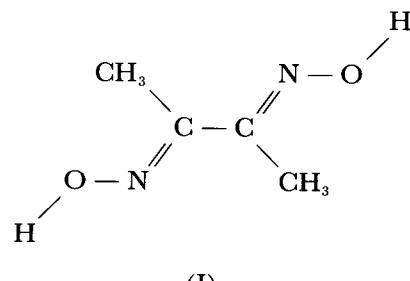
Free energies of monatomic ions can be correlated fairly well with radius and charge. If ionic radii suggested by the partial molal volumes are used, the extrapolated free energy for an ion of infinite size is zero. This is in agreement with the Born model for ion solvation.

### Neutron Diffraction by Single Crystals

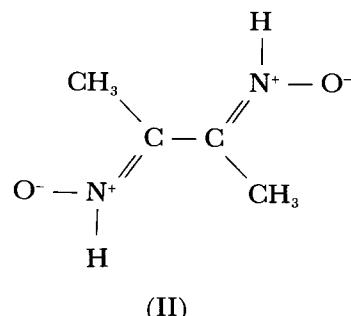
With increasing use of a single-crystal neutron diffractometer, the work on neutron diffraction has been expanded to include the location of hydrogen atoms in complex crystal structures. Two compounds have been studied within the past year: dimethylglyoxime and sulfamic acid. Previous studies of both molecules by the x-ray diffraction method resulted in structure determinations that were complete except for determination of hydrogen atom locations. This information was lacking because of the unfavorable ratio of heavy atom to hydrogen atom scattering factors.

In dimethylglyoxime, the planar molecules are linked together in sheets by hydrogen bonds between nitrogen and oxygen atoms, in which the

hydrogen could be chemically bonded to either the nitrogen or the oxygen atom. The two possible molecular structures may be written

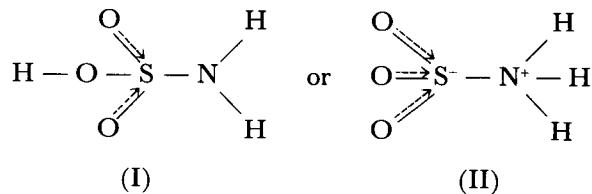


or



A single-crystal neutron diffraction study has indicated unambiguously that structure (I) is correct and has further led to determination of accurate positions for the hydrogens in the methyl group.

Similarly, sulfamic acid may be written



By the use of single-crystal neutron diffraction techniques, structure (II), the zwitterion structure, has been shown to be correct. The molecule has approximately threefold symmetry with some distortion of the  $\text{NH}_3$  group to allow more favorable hydrogen bonding.

For both molecules, accurate values for bond lengths and angles have been obtained, and there are no major departures from expected values. The S-N single bond length of 1.764 Å has not

previously been determined with accuracy. The  $\text{N} \cdots \text{H} \cdots \text{O}$  hydrogen bond angles in both molecules depart from linearity by 10 to 20°.

In connection with the single-crystal work, extensive use has been made of the IBM 704 computer at the New York University Computing Facility. A library of programs for routine crystallographic calculations and data reduction has been accumulated.

In another application of single-crystal neutron diffraction, magnetic scattering has made possible the investigation of the nature of the second-order phase transition which occurs in magnetite ( $\text{Fe}_3\text{O}_4$ ) at 119° K. The great decrease in electrical conductivity which accompanies this transition led Verwey and co-workers to postulate that the transition involves an ordering of ferric and ferrous ions which have a random arrangement above the transition temperature. The problem of verification of this ordering, nearly impossible of solution by x-ray diffraction, is especially susceptible to attack by neutron diffraction because of the difference between the magnetic scattering cross sections of the ferric and ferrous ions. Measurements on single crystals have unambiguously established the validity of the Verwey ordering scheme.

Magnetite above the transition temperature has cubic symmetry, the structure being of the inverse spinel type with a random arrangement of ferrous

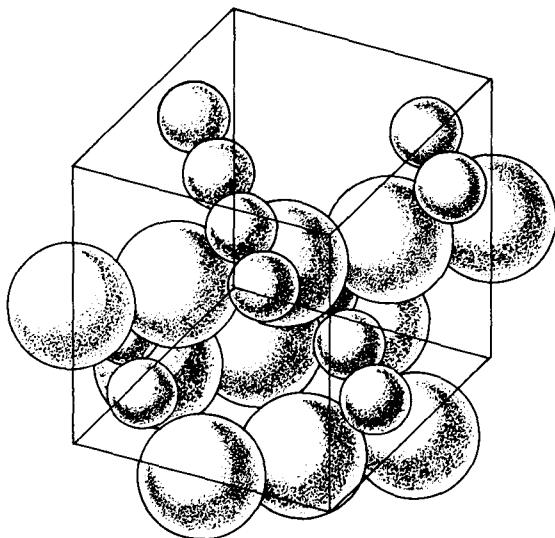


Figure 6. The arrangement of ferric ions (small spheres) and ferrous ions (large spheres) in magnetite, at temperatures below 119° K.

and ferric ions in the octahedrally coordinated sites. At the transition it passes over to orthorhombic symmetry. Diffracted intensities were measured in the absence of a magnetic field and with a magnetic field applied in various directions. The results, including the observation of the (002) reflection, forbidden in the cubic phase, indicate the following:

If the orthorhombic axes are labeled such that the magnetic anisotropy energies are in the order  $E_a > E_b > E_c$ , i.e.,  $c$  is the direction of easy magnetization, then

- 1) the octahedral ferric ions lie in rows parallel to  $a$ ;
- 2) the octahedral ferrous ions lie in rows parallel to  $b$ ;
- 3) the magnetic moments of all ions are parallel or antiparallel to  $c$ ; and
- 4) the  $a$  axis is longer than the  $b$  axis.

Figure 6 indicates the arrangement of the ions in the octahedral sites relative to the original cubic cell. The large spheres represent ferrous ions, the small spheres ferric ions.

#### Neutron Diffraction by Magnetic Crystals

In the last few years Russian crystallographers and mathematicians have worked out the extension of the 230 symmetry space groups to include operations of antisymmetry acting on objects endowed with black and white color. Antisymmetry operations combine ordinary symmetry operations with a change in color. A total of 1651 groups, called the Shubnikov groups, has been enumerated: 230 uncolored (classical), 1191 black-and-white, and 230 gray (black and white objects superposed). The uncolored and black-and-white groups may, with suitable reinterpretation, be used to describe structures possessing aligned but not necessarily collinear magnetic moments; the gray groups are applicable to paramagnetic crystals. The change of color that characterizes operations of antisymmetry in the black-and-white groups is replaced by the reversal of spin direction,  $R$ , to be combined with the corresponding symmetry operation. The operation of identity combined with  $R$  is allowed only in the gray groups; it does not occur in black-and-white groups. The transformation properties of magnetic moments differ from those of black-and-white objects so that the positions available in a given space group may be different for the two cases. The effects of

OPERATIONS OF THE FIRST KIND		OPERATIONS OF THE SECOND KIND		
TRANSLATION - $t$	ROTATION - 2	INVERSION - $\bar{1}$	ROTATORY INVERSION - $\bar{4}$	REFLECTION - $m$
ANTITRANSLATION - $t'$	ANTIROTATION - $2'$	ANTIINVERSION - $\bar{1}'$	ANTIROTATORY INVERSION - $\bar{4}'$	ANTIREFLECTION - $m'$

Figure 7. The effects of symmetry and antisymmetry operations on magnetic moments.

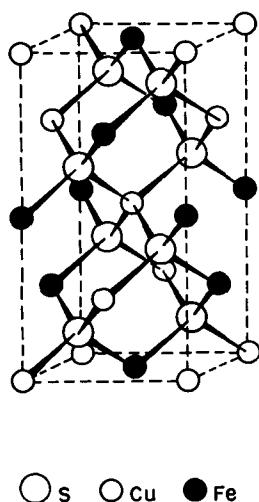


Figure 8. The crystal structure of chalcopyrite,  $\text{CuFeS}_2$ .

symmetry and antisymmetry operations are summarized in Figure 7.

The problem of placing magnetic moments in a ferro- or antiferromagnetic crystal structure previously determined by x-rays can be attacked by a trial-and-error procedure that ignores the symmetry properties of magnetic moments. For a cell containing  $N$  atoms this requires, in principle, the refinement of  $3N$  positional parameters quite apart from the directional parameters of the magnetic moments. This procedure is unsatisfactory for complex structures and may even present difficulties in simple structures.

Alternatively, since magnetic moments are subject to symmetry and antisymmetry operations, a systematic procedure can be followed by starting with a spin distribution that obeys the highest possible symmetry compatible with the chemical space group and the neutron diffraction data. The latter, if different from the x-ray data, are used to determine the size of the magnetic cell, its symmetry, and its diffraction aspect. The chemical space group, or its most symmetrical subgroup compatible with the magnetic diffraction aspect, is the first space group to be tried. If the agreement of structure factors is not satisfactory, the next step is to try the antigroups (black-and-white) derivable from the chemical space group and permitted by the magnetic diffraction aspect. The next lower space group permitted by the diffraction aspect is tried only after these possibilities have been ruled out. Additional positional parameters are thus in-

troduced which may be varied to improve the agreement within the limits of accuracy of the x-ray determination. This procedure has been applied to chalcopyrite,  $\text{CuFeS}_2$ , which is discussed below.

Previous studies of antiferromagnetic compounds involving tetrahedral coordination of magnetic atoms about an anion have shown that the local magnetic structure is consistent with an indirect exchange coupling mechanism. Additional examples of such structures were sought in order to study the effect of tetrahedral bonding and covalency on the interaction of magnetic atoms. The appearance of antiferromagnetism in the zinc blende and wurtzite forms of  $\text{MnS}$  suggested that  $\text{CuFeS}_2$  might also prove to be antiferromagnetic.

Measurements of the magnetic susceptibility were carried out by the Gouy method. Between 77°K and room temperature the susceptibility was found to be low, with a small positive temperature coefficient. Above room temperature the susceptibility increased more rapidly with increase in temperature but was not completely reversible. The dependence on thermal history may have been caused by chemical reaction with impurities present in the sample. While the evidence for antiferromagnetism obtained from these measurements is not entirely conclusive, further confirmation was obtained from the presence of strong additional reflections in the neutron powder and single-crystal diffraction patterns.

The results of x-ray and neutron measurements indicate that the chemical structure is that proposed by Pauling and Brockway (Figure 8). The possible existence of a second chalcopyrite modification in nature, suggested by conflicting results on material of Japanese origin, is ruled out, since specimens from both Ugo, Japan, and Joplin, Mo., are found to have the same structure. The chemical space group  $I\bar{4}2d$  holds also for the magnetic structure, in which the two irons tetrahedrally bonded to a common sulfur atom have antiparallel spins directed along the tetragonal axis. A value of  $3.85 \mu_B$  is found for the iron moment ( $0 \pm 0.2 \mu_B$  for copper). The configuration of iron moments relative to the symmetry elements of  $I\bar{4}2d$  is shown in Figure 9.

In its magnetic structure  $\text{CuFeS}_2$  closely resembles the zinc blende form of  $\text{MnS}$ . If, in the latter structure, those magnetic moments corresponding to the copper sites in  $\text{CuFeS}_2$  are deleted, the antiferromagnetic structure observed for  $\text{CuFeS}_2$  is ob-

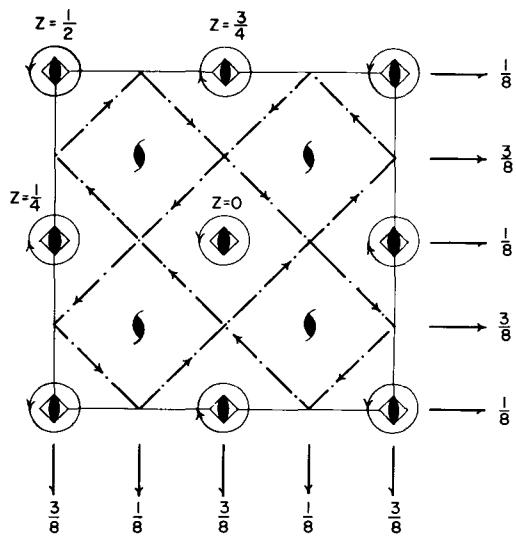


Figure 9. The configuration of the magnetic moments of iron atoms in chalcopyrite,  $\text{CuFeS}_2$ .

tained. In  $\text{MnS}$ , each sulfur is tetrahedrally bonded to four metal atoms which are pairwise antiparallel, and it cannot be said *a priori* whether the indirect exchange interaction between a pair of metal atoms is ferromagnetic or antiferromagnetic. The evidence provided by the magnetic structure of  $\text{CuFeS}_2$  strongly suggests that this interaction is antiferromagnetic.

#### Internal Consistency of the Heisenberg-Dirac Model for Antiferromagnetism

The Heisenberg-Dirac model for ferromagnetism and antiferromagnetism assumes an interaction between any two atoms  $i$  and  $j$  of the crystal which is of the form

$$V_{ij} = -2J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j.$$

Here  $J_{ij}$  is an exchange interaction and  $\mathbf{S}_i$  and  $\mathbf{S}_j$  are the spin operators for the respective atoms. It has not been possible to obtain exact solutions for this model, but various high temperature approximations have been developed which may be used to predict the transition temperature and the physical properties above the transition temperature. The results so obtained are in good qualitative agreement with experiment, but no very discriminating tests of the general consistency of the theory have been made, principally because the theoretical treatments have usually been limited to the case of spin  $1/2$  per atom and to nearest-neighbor interactions only, while practically all

Table 8

#### High Temperature Values of Nearest-Neighbor Exchange Interaction

Compound	Values of $J/k$		
	From $T_N$	From $\chi(T_N)$	From $C_p$
$\text{MnF}_2$	1.80	1.97	1.73
$\text{FeF}_2$	2.88	2.98	3.01
$\text{LaFeO}_3$	26.2	28.9	—
$\text{LaCrO}_3$	26.0	20.0	—

the experimental data are for more complex systems. In an effort partially to bridge this gap and thus make possible sensible comparisons between theory and experiment, the Bethe-Peierls-Weiss method has been extended to calculations of the high temperature properties of antiferromagnets having spin per atom greater than  $1/2$ ; the nearest-neighbor restriction has been retained, however. One result of this work is an expression showing how the susceptibility of the Néel temperature depends on the nearest-neighbor exchange interaction,  $J$ ; the relation between the Néel temperature itself and  $J$  has previously been given by Brown and Leitinger. Thus, if  $T_N$  and  $\chi(T_N)$  are measured, the experimental results can be combined with the theoretical relations to give two independent values of  $J$ ; the two values should agree if the theory is internally consistent.

There is only a limited number of antiferromagnetic systems which seem to satisfy the nearest-neighbor restriction and for which the requisite measurements have been made. Results are shown for four compounds in Table 8. The figures in the fourth column are independent determinations of  $J/k$  made by Hofmann, Paskin, Tauer, and Weiss from an analysis of specific heat data. The discrepancies between the values in columns 2 and 3 are of the order of 10% and may be attributed to four possible sources: 1) a deficiency of the Heisenberg-Dirac model, 2) a deficiency of the Bethe-Peierls-Weiss approximation, 3) effects of second-nearest and more distant neighbors, and 4) experimental error. There is no way of separating the contribution of each to the total discrepancy, and it is not unlikely that all four may be significant. In any event, the over-all agreement is reasonably good and suggests that the Heisenberg-Dirac model can

be used with confidence to discuss quantitatively the high temperature properties of antiferromagnets, although no great precision should be expected.

## MISCELLANEOUS SUBJECTS

### Stationary Damping Theory

It is well known that ordinary perturbation theory is not adequate for the task of finding the stationary states of systems in which radiative damping phenomena can occur. Such a system is an atom in interaction with radiation near one of its resonance frequencies. The difficulty arises from the infinite degeneracy of the problem, and from the fact that there is not always a one-to-one correspondence between perturbed and unperturbed states. It can happen that some of the unperturbed states have no exact stationary state corresponding to them. An example of this is an excited state of an atom or molecule.

Simple problems of this type, such as resonance scattering by a single atom, can be solved without difficulty; the general case, however, is not so easy. For instance, the one-atom resonance scattering can be represented by saying that the absorption is "damped" by the decay of the excited state, and the denominator of the expression for the scattering amplitude contains an imaginary term which is essentially the inverse lifetime of the excited state. However, if many identical atoms are present, the excited state can decay by exciting another atom as well as by radiation, so that the "lifetime" depends on the presence of other atoms and on their lifetimes, etc. This can become quite complicated.

A general formalism has been developed for dealing with such problems by using some new operators, the "damping operators." The method includes a criterion for deciding whether or not a given unperturbed state has an exact stationary state corresponding to it.

A simple example is the complex refractive index of a gas. Here the nonresonance transitions can be eliminated by the transformation of Arnous and Bleuler. Formally exact solutions can be obtained for frequencies near resonance, corresponding to attenuated waves with complex refractive index given by

$$(n^2 - 1)(\nu) = - \frac{4\pi N}{V} \frac{\nu_0^2}{\nu^2} \frac{M^2}{\hbar(\nu - \nu_0) - \langle i | D(\hbar\nu) | i \rangle}.$$

Here  $| i \rangle$  is the state in which one atom of the gas has been excited,  $M^2$  is the square of the appropriate dipole matrix element,  $\nu_0$  is the resonance angular frequency,  $\langle i | D(\hbar\nu) | i \rangle$  is a matrix element of a "damping operator," and  $N/V$  is the number of atoms per unit volume. For simplicity, small corrections due to the nonresonance transitions have been omitted from the preceding equation. If interatomic interactions are neglected, the  $D$  matrix element in the denominator is essentially independent of  $\nu$  and is equal to  $-i\hbar$  times the natural line breadth. If interatomic interactions are included, the dependence of  $D$  on  $\nu$  changes the shape of the absorption curve from the simple Lorentzian form and gives an effective width of the same order of magnitude as the "resonance broadening" calculated elsewhere. An exact calculation of this effect, however, has not yet been carried out because of mathematical difficulties.

### Ion Exchange Equilibria

Ion exchange equilibria on single resin beads have been studied for the silver ion - hydrogen ion system by the use of radioactive silver. These measurements show that for some commercial resins a large bead-to-bead variation exists. This heterogeneity effect has been studied theoretically by treating an ion exchanger in formal analogy with a heterogeneous surface. It is found that experimental data can be represented almost equally well by a system with two different exchange constants or by a system with practically an infinite number of constants (i.e., each site has its own constant). In this latter system there is a nearly Gaussian distribution of the number of sites with respect to the free energy of reaction.

Equilibrium data alone cannot be used to distinguish between the system with a few exchange constants and the system with an infinite number of constants. Hence, autoradiography is being used in an attempt to obtain additional information about the distribution of ions in the resin phase.

A method of determining partial molar volumes of resin species has been developed and applied to the potassium-tetramethylammonium system studied by Gregor et al. The volume change upon mixing is found to be ideal over the large range of 1 to 18 molal (total average concentration of resin). Measurements on the silver ion - hydrogen ion system also show an ideal change of volume upon mixing.

### **Chemical Composition of Archaeological Artifacts**

Studies correlating the provenance of archaeological artifacts with their chemical composition have been continued by means of spectroscopic analysis. Among the pottery specimens investigated, a group of stamped amphorae handles excavated at the Athens agora showed the greatest degree of correlation and revealed some interesting historical information. The jars had been used to transport wine to the Athens market, and the handles bore stamps indicating the origin of the wine. Eleven of twelve handles identified as having come from Knidos in Asia Minor had similar patterns of constituents. The twelfth differed markedly from these but agreed closely with three specimens identified as having originated at Rhodes. After further examination by the archaeologist supplying the specimen, it was concluded

that the stamp on the twelfth handle clearly indicated Knidos as the source of the wine but corresponded to a period when Knidos was ruled by Rhodes.

Each sample in a group of more than 100 specimens of ancient glass obtained from private and museum collections is being spectroscopically analyzed for 27 elements. The specimens represent nearly all major ancient sites of glass manufacture in the Near East, Africa, and Europe. The types of glass are all much alike in composition; however, certain major discriminations can be made. For example, there are differences between the earliest Egyptian glass, that produced during the Roman period, and that of the Islamic period. Although specimens of Islamic glass produced throughout the near East are similar, certain trace constituents seem to distinguish the glass produced in Asia Minor from that produced in Egypt.

# Nuclear Engineering

The research activities in nuclear engineering have been chiefly concerned with the development of new reactor systems or system components to further the economic development of nuclear energy. More than half the total effort was devoted to research on the Liquid Metal Fuel Reactor (LMFR) during the past year. The Babcock & Wilcox Company, under contract with the Atomic Energy Commission, has undertaken the design and construction of a Liquid Metal Fuel Reactor Experiment (LMFRE). The Laboratory's increased responsibility for research and development directly required for the design of the LMFRE has necessitated an increase in personnel and in magnitude of effort.

Activities other than those involved in the LMFR have included studies of reactor physics problems, fuel development and processing, waste processing and disposal, radiation chemistry, heat transfer, and radioisotope development.

## LIQUID METAL FUEL REACTOR

The use of liquid metals as reactor fuels has been under investigation at Brookhaven for several years. The search continues for satisfactory solutions to corrosion and mass transfer problems involving the liquid metal fuel and its containing vessel.  $U^{235}$  dissolved in molten bismuth together with such inhibitors as zirconium and magnesium constitutes the fuel. Graphite will probably be used as a moderator. The container material in most experiments has been a low chrome steel. It is hoped to maintain a low concentration of reactor poisons by the continuous removal of fission products from the fuel. The possibility of developing a blanket material in liquid form for breeding purposes has also been under investigation. The progress made during the past year in obtaining solutions to the many problems involved is summarized below.

### Reactor Physics

A Neutron Source Reactor has been constructed to provide neutrons for the LMFR ex-

ponential studies. The reactor design is similar to that of the Medical Research Reactor, having been developed from the critical experiments on that facility. The core is formed of MTR-type enriched uranium fuel elements, aluminum clad. The moderator and coolant are demineralized water, and the core is reflected by graphite on all sides. A summary of the reactor characteristics is given in Table 1.

The horizontal thermal column for LMFR exponential experiments is adjustable in cross section up to  $48 \times 48$  in., and in length from 16 to 40 in. In addition, a larger thermal column cross section is available by removing several shielding blocks. The vertical thermal column consists of a 78-in.-diameter water tank that rests on the top of the graphite reflector structure.

The presence of the top thermal column precludes the normal vertical control arrangement. Instead, the control rods (four in number) operate horizontally in channels which pass through the

Table 1

Neutron Source Reactor	
Core	MTR type: 16 fuel elements, each containing 140 g $U^{235}$ .
Moderator and coolant	Demineralized water, circulated in a closed system.
Reflector	AGHT and AGR graphite.
Heat exchangers	Shell and tube type, water to water.
Shielding	Heavy concrete, 20 in.; ordinary cement blocks, 48 in.
Maximum power	100 kw.
Experimental facilities	Horizontal thermal column (graphite). Vertical thermal column (graphite and water). Three $1 \times 1$ -in. holes through core.
Maximum flux at thermal columns	$10^{10}$ Neutrons/cm <sup>2</sup> -sec.
Maximum flux in core holes	$10^{12}$ Neutrons/cm <sup>2</sup> -sec.

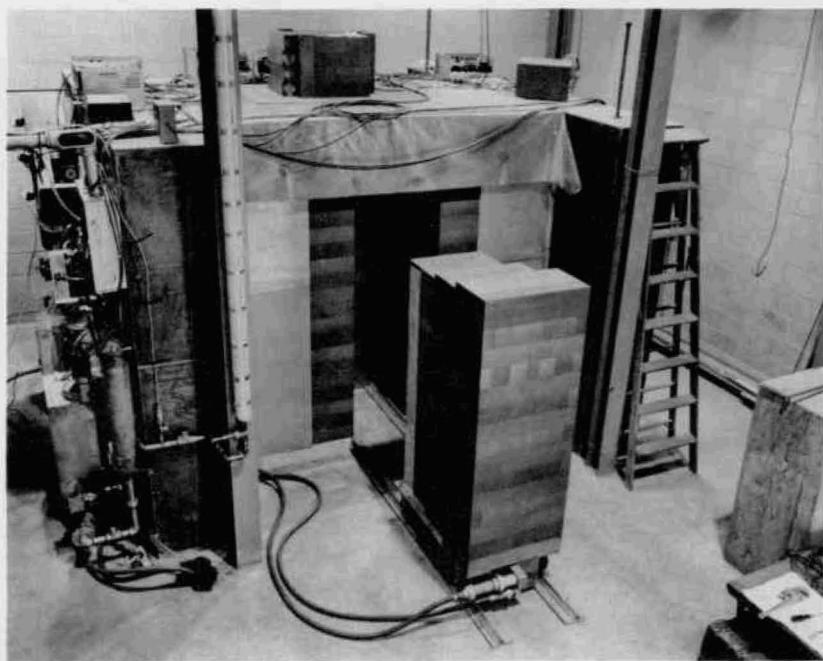


Figure 1. The Neutron Source Reactor is shown during start-up experiments. One of the core tank carriages has been pulled out of the graphite reflector structure to permit changes in the loading.

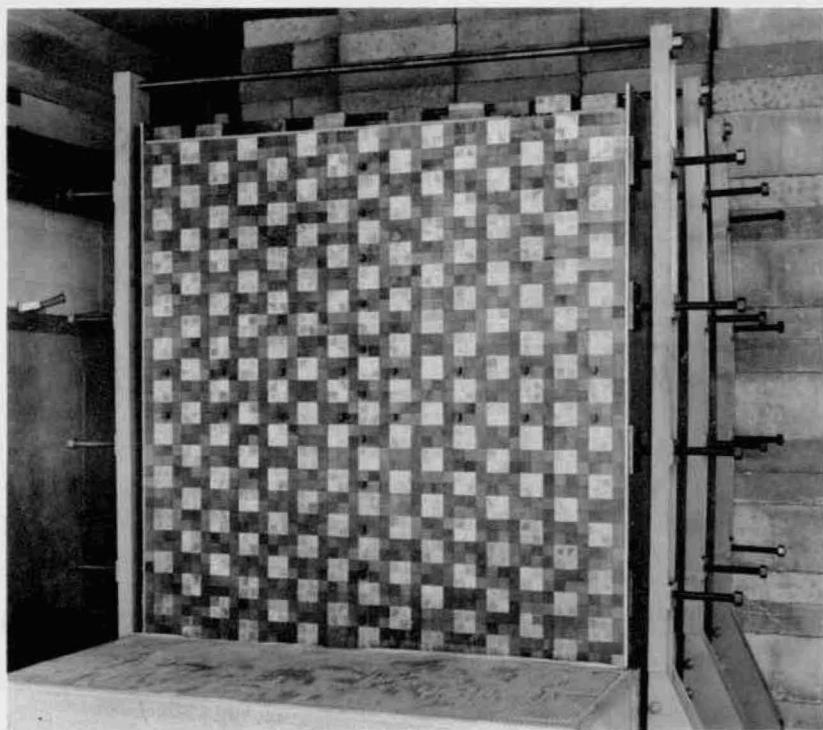


Figure 2. Graphite-bismuth-aluminum array for the measurement of the diffusion length in a volume ratio of 2 graphites to 1 bismuth with a 2-in. fuel module.

center of the core. The core vessel consists of two tanks, separated by a 2-in. graphite septum, with a separate water circulation system for each tank.

The shielding has been arranged to give maximum flexibility in the exponential experiments. A comparatively thin shield of heavy concrete blocks reduces the room-reflected neutron background to an acceptable level during the operation of the reactor, and provides sufficient shielding for the shutdown activity. A second shield of ordinary cement blocks outside the reactor room serves as the biological shield during operation.

Since the LMFR exponential experiments will not require the full design power of 100 kw until breeding blanket studies are started, authorization was sought from the AEC to operate the Neutron Source Reactor at 1-kw power. This authorization was given on May 19, 1958, and the reactor was brought critical on that date. Start-up experiments on control rod worths and various reactivity coefficients occupied the remainder of the year. Figure 1 shows the reactor during the start-up experiments.

A series of measurements of neutron slowing down and diffusion parameters in various LMFR core materials has been made as a preparatory step in the LMFR exponential experiments. The neutron thermal diffusion length has been measured in graphite (specifically, the graphite to be used in the exponential experiments), bismuth, and several mixed graphite-bismuth-aluminum arrays. The arrays include a volume ratio of 2 graphites to 1 bismuth, with 1-in. and 2-in. fuel modules (Figure 2). Values of the transport mean free path have been obtained by the extrapolation distance method as a by-product of the diffusion length in each of the above core arrangements. The transport mean free path has also been measured in graphite by an extensive series of experiments involving the poisoning of the graphite array with various amounts of pure copper.

The neutron age to indium resonance energy has been measured in graphite and in the mixed arrays 2 graphites - 1 bismuth, with 1-in. and 2-in. fuel modules. The results of these measurements have been reported in a paper and supplement submitted to the second Geneva Conference. The outstanding problems remaining in connection with diffusion measurements are the determination of the transport mean free path in bismuth by the poison method, and the resolution of questions involving an apparent anisotropy in the

diffusion of neutrons parallel and antiparallel to the extrusion axis in graphite.

A method of activation analysis for chlorine content in bismuth in trace amounts was developed and applied to the bismuth to be used in the LMFR exponential experiments. The chlorine content was found to be about 1 ppm by weight, an amount too small to influence the exponential measurements.

A simplified calculation of the transfer function for LMFR systems was coded for the BNL Remington Rand computer. The results show a tendency for resonances to occur at frequencies in the range of bismuth circulation frequencies. In an effort to improve the calculation, the problem of heat transfer between fuel and graphite in the LMFR was explored on the small analogue computer at the Cosmotron. The computer accuracy for this problem is poor, however, and a much improved analogue system will be needed to obtain credible results.

#### Fission Product Removal Chemistry

Problems connected with the removal of fission products from liquid metal fuel and with the interaction of the fuel with its environment have continued to provide the stimulus for most of the LMFR chemical research. Of the fission products, the group extractable by fused salts (the FPS group) has received the most attention. The FPS removal process has been brought closer to practical application, and further information has been obtained about the nature of the salt and metal phases involved and the mechanism of their interaction. For the other groups of fission products, the volatile (FPV) and the relatively noble (FPN), a separation process cannot yet be prescribed, but much has been learned about their behavior. To the crucial question of whether fission products and other solutes in Bi will be adsorbed on the graphite and steel surfaces to which the fuel will be exposed, small-scale experiments are beginning to indicate the answer.

Perhaps the most important development in the technology of the salt process has been the application of successive oxidation and reduction steps to a single salt-metal system, in a batch operation at the kilogram level.

In any practical extraction process, a little U can be expected to transfer to the salt phase along with the bulk of the FPS fission products. It is proposed that this U be recovered from the salt

by scrubbing with Bi containing more Mg than the fuel. This reduction step has now been proved feasible. In the experiments, Bi containing Mg, Zr, and U was equilibrated with the fused triple eutectic  $MgCl_2$ - $NaCl$ - $KCl$  in a 2-liter bench contactor. When the composition of each phase had stabilized, a quantity of  $BiCl_3$  was added. If this was less than equivalent to the total Mg in the metal phase, only Mg reacted and the U concentration remained constant after some initial fluctuations. If, however, there was more than enough  $BiCl_3$  for the Mg, the U also reacted and was transferred as the chloride to the salt phase. This reaction was completely reversible upon the addition of fresh Mg to the metal. The bench contactor results were, surprisingly, more favorable in this respect than those of gram-scale equilibrations of specifically purified materials.

Information on the distribution of rare earth tracers as a function of Mg concentration was also obtained from the bench contactor systems. Earlier results were in general confirmed, and separation factors of well over 100 may be expected between the rare earths and U; separation of the alkali metal and alkaline earth fission products should be even more efficient. This behavior will be made use of in practice by contacting fuel withdrawn from the reactor with fused salt in some countercurrent system, either a column or a series of equilibrium stages.

#### Extraction Kinetics Pilot Plant

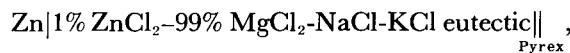
In the extraction kinetics pilot plant, the current plan is to use columns, the salt flowing upward as the continuous phase. In some small-scale experiments, single drops of Bi containing Sm were dropped through columns of molten salt, and the results showed that the Sm was quickly extracted from the Bi. The very favorable kinetics obtained in these experiments augur well for the use of extraction columns. The electrochemical behavior of such extraction columns has been studied in experiments in which Hg and water were substituted for Bi and salt, with different chemical reactions taking place at different oxidation-reduction potentials.

The final testing of the extraction columns on a nonradioactive basis will be done in Loop N, a large, complex facility being built specifically for this purpose at the O.G. Kelley Company's plant in Boston. This facility will provide continuous streams of salt and fuel to the test columns.

#### Process Instrumentation

In a continuous process it will be important to know the composition of each phase at each stage at all times. If possible, instruments should be developed that will give continuous signals indicating and recording the important process variables. In this connection, already initiated research programs to explore the possibilities of absorption spectroscopy, measurement of electrode potential, and polarography were continued and amplified. Spectroscopic results obtained by Professor H.M. Haendler of the University of New Hampshire, working under a research subcontract, have confirmed that spectroscopy can be a sensitive method for the determination of  $UCl_3$  in salt. For example, a salt mixture containing 0.061 wt %  $UCl_3$  had an absorbance (over that of the solvent) of 1.43 at 309  $m\mu$  for a 1-cm light path. In connection with the spectroscopy program, studies have been made of the effect of gamma radiation on silica cell windows, since the actual process salt will be extremely radioactive. The coloration resulting when cells were irradiated at 425°C or below was completely removed by annealing at 500° for a short time, and it now seems likely that radiation damage will not preclude the use of spectrophotometric methods.

The electric potential at the salt-metal interface is related to the composition of the two phases. In the LMFR system it will be chiefly responsive to the Mg/ $MgCl_2$  ratio; in other words, since the  $MgCl_2$  concentration will remain sensibly constant, it measures the concentration of Mg in the Bi phase. The reference electrode developed to measure this potential, which consists of the half-cell



has been tested in equipment of the bench contactor type that contained salt and Bi phases. The emf measured between the Bi and the reference half-cell was in fact an accurate index of the amount of Mg in the Bi. On this principle a direct-reading instrument is being constructed in which temperature variations will be automatically compensated.

The application of polarography to fused salts has been further developed during the past year. The performance of the dropping Bi electrode was much improved by lengthening the Pyrex capillary tube from which the drops fall until drop

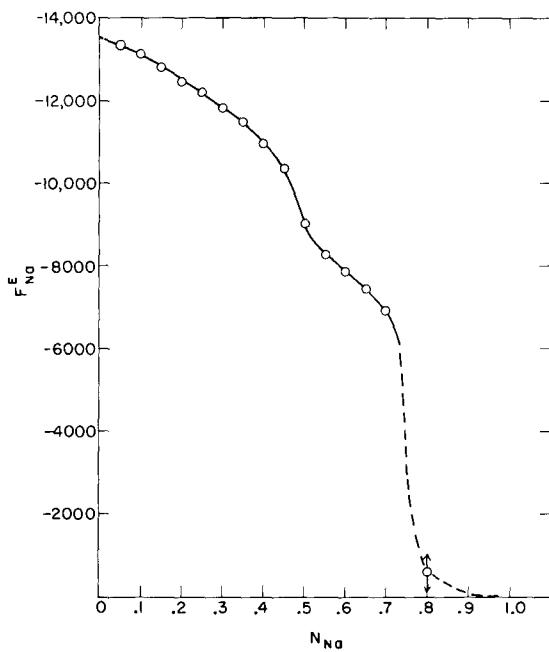


Figure 3. Partial molar excess free energy of mixing of Na in the system Na-Bi.

times as long as 5 sec were obtained. Analysis of the results showed good agreement with the Ilkovic equation.

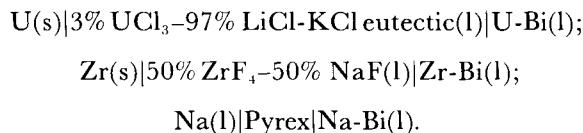
Reasonable values were found for the diffusion coefficient of the reducible species: e.g.,  $D = (2.0 \pm 0.4) \times 10^{-5}$  cm<sup>2</sup>/sec for PbCl<sub>2</sub> in the LiCl-KCl eutectic at 450°C. It was also possible to determine the concentrations of PbCl<sub>2</sub> to an accuracy of better than 10% in the range 0 to 1000 ppm. Attempts to analyze for U in this way were unsuccessful; the supporting electrolyte began to decompose before the U<sup>+++</sup> → U wave appeared.

A related research program is being carried on at Rensselaer Polytechnic Institute by Drs. Oster- young and Hill, who have a research subcontract with the Laboratory. They also are obtaining polarograms on fused salt mixtures, with the difference that theirs is a Pt microelectrode instead of dropping Bi. According to their preliminary results a U reduction wave can be obtained in this system.

#### Thermodynamics of Liquid Metals and Fused Salts

Much of the quantitative thermodynamic information needed for a complete understanding of the salt extraction process is still nonexistent. To fill this gap, part of the past year's LMFR program has consisted, as in previous years, of

measurements of the free energies of mixing of various solute metals in Bi and of salts in salt mixtures. For both types of system, use has been made of emf measurements on appropriate galvanic cells. With metals, electrode concentration cells such as the following were set up:



In these cells the emf measured the partial molar free energy of the solute in the alloy electrode, the Bi taking no part in the cell reaction. The results on the Na system, measured over a wide range of concentration, are shown in Figure 3. Large deviations from ideality were also found in the other systems. For example, the following activity coefficients were found at 700°C:

SOLUTE MOLE FRACTION IN BI ACTIVITY COEFFICIENT,  $\gamma$

U	0.0068	$4.4 \times 10^{-4}$
	.0042	$4.5 \times 10^{-4}$
Zr	.0116	$6.5 \times 10^{-3}$
	.0194	$6.2 \times 10^{-3}$

For research on salts a similar type of cell was used:



From this, the free energy of mixing of MgCl<sub>2</sub> in the salt mixture can be obtained. Figure 4 presents some results for this system in graphical form. Similar data were obtained for the systems MgCl<sub>2</sub>-NaCl, MgCl<sub>2</sub>-NaCl-KCl, and CeCl<sub>3</sub>-KCl.

#### Technology of Fused Salts

Experiments and tests at all levels were carried out to determine how to handle fused chloride mixtures on an engineering scale. Corrosion studies have been carried out in static, dynamic, and circulating experiments. The dynamic tests were conducted both with a temperature difference and in the presence of U-Bi fuel, while the circulating loop tests were conducted with a temperature difference only. From results obtained to date, it seems that for cases in which the fuel and the ternary eutectic are present together the most usable materials of construction are the low chrome steels. For the straight ternary salt, 347 stainless steel appears to be satisfactory. For salt mixtures containing more than 1% BiCl<sub>3</sub>, the

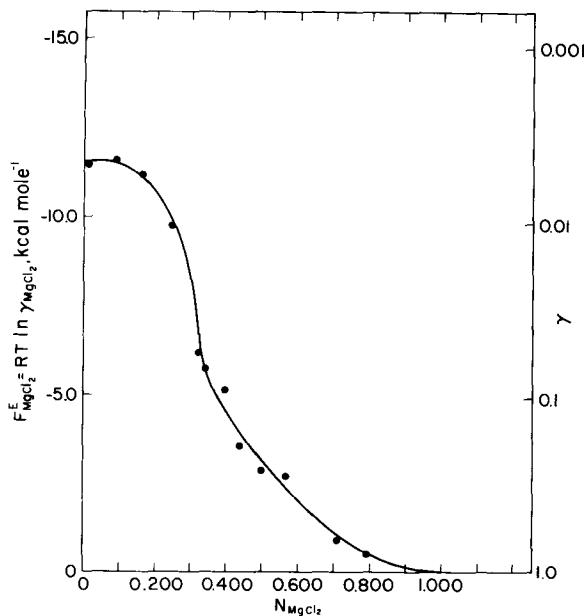


Figure 4. Excess partial molar free energy of  $\text{MgCl}_2$ ,  $F_{\text{MgCl}_2}^E$ , in  $\text{MgCl}_2\text{-KCl}$  melts at  $800^\circ\text{C}$ .

usual oxidant, steels seem to be inadequate but molybdenum appears promising.

Loop M, an isothermal pumped salt loop made of 347 stainless steel, was operated for part of the year to test mechanical components for fused salt service. Valves, pumps, and instrumentation devices for handling flowing liquids were tested. Most of the mechanical components for Loop N, the facility for testing the effectiveness of extraction columns for salt treatment of the fuel, were tested in advance in Loop M. The fused salt technology is considered to have advanced to the point where small engineering systems can be built with reasonable expectation of satisfactory operation.

Of those fission products which are neither volatile nor more readily oxidized than U, the FPN's, the most important are the elements that are more noble than Bi and hence cannot be slagged out by an oxidant. They are Ru, Rh, Pd, Mo, Te, and maybe Tc. The results of a few experiments on Ru-Bi systems indicated that extraction with liquid Zn might remove some of the FPN's. Since manpower and space were lacking at Brookhaven, the American Smelting and Refining Company was commissioned to conduct research, under subcontract, on Zn processes. It has been found that the addition of as little as 0.5% Zn to Bi containing Ru, Rh, Pd, and Te is effective in

removing these solutes. They react with Zn to form intermetallic compounds that become insoluble as the temperature is lowered and can be filtered out or skimmed off.

### Polonium

The behavior of Po in the various processes to which LMFR fuel is subjected is of interest because of the element's great toxicity. In salt extraction, tests have shown that all but  $\approx 0.1\%$  of the total Po remains in the Bi phase, regardless of the oxidation potential of the system. What little does go into the salt phase apparently is transferred by some mechanism other than oxidation. When Bi containing Po was equilibrated with liquid Zn, Po again remained almost entirely in the Bi phase.

### Adsorption Effects

The fuel of an LMFR will be exposed to solid surfaces on which some of the solutes may well become concentrated, either by reaction or adsorption. Investigation of some of these surface effects has been initiated. Bi containing Sm tracer was contacted with graphite surfaces under various conditions, and the amount of Sm remaining on the graphite was determined. In general, only a few tenths of a percent was retained.

### Volatile Fission Product Removal

In order to maintain low concentrations of neutron poisons in an LMFR, it is desirable to remove continuously certain groups of fission products. In this respect, the FPV's (Xe and Kr isotopes) are the most important because of the very high cross section of  $\text{Xe}^{135}$ . The degree to which Xe may be removed from LMFR fuels depends on: 1) the solubility of Xe in U-Bi fuel, 2) the extents to which Xe and its precursor I tend to be adsorbed by and absorbed in the unclad graphite core, and 3) the kinetics of Xe desorption in a particular type of degassing device. During the past year a research program to elucidate these three points has been under way. The results thus far may be summarized as follows:

1) Preliminary results on the measurement of Xe solubility in U-Bi fuel (1000 ppm U, 350 ppm Mg, and 250 ppm Zr) at  $500^\circ\text{C}$  and under a pressure of about  $1\frac{1}{3}$  atmospheres is in the neighborhood of  $10^{-8}$  mole fraction.

2) Both Xe and I are adsorbed to a very considerable degree on high density reactor-grade

graphites. In in-pile experiments,  $\approx 99\%$  of the I and 90% of the Xe in the U-Bi fuel are found in the graphite walls of the capsules used in these experiments. At 500°C the penetration rates of Xe and I are such that the front of a given concentration advances through the graphite at the approximate rate of 0.2 mm/hr when the concentration of these nuclides in the U-Bi fuel is about  $10^{13}$  atoms/g. Concentrations of  $10^{12}$  atoms Xe<sup>133</sup> and  $10^{14}$  atoms I<sup>131</sup> per g graphite have been measured.

The upper limit of Xe absorbed by graphite has been determined in a separate series of experiments (absorption from gas phase at 610 mm and 500°C) to be  $10^{17}$  atoms/g graphite. The work on gas phase absorption is continuing in order to establish isotherms in the 500° to 800°C range and at pressures up to one atmosphere. An indirect result of this work is the indication that volatile iodine compounds (possibly BiI<sub>3</sub> or Bi<sub>x</sub>I<sub>y</sub>) are formed in LMFR fuel. Work in progress should result in further clarification of this point.

3) A bench-scale facility for studying the kinetics of Xe removal and the efficiencies of several types of degassers has been designed, and most of the construction work has been completed. This facility is expected to be in operation by the end of 1958.

#### Loops for Component and Corrosion Testing

In addition to those already mentioned, several loops were being built, revised, or operated during the year. Among these were three loops designed principally for the testing and development of components. Loop H-1, a multicomponent, forced circulation Bi loop, was operated periodically throughout the year. A sister loop, H-2, is still under construction. A very large mechanical component test loop (4-in. loop) was under construction during the year. This facility will be housed in a special building. Its main purpose is to provide a full-scale prototype test of various components to be used in LMFRE No. 1, including pumps, valves, pipe, pipe fittings, vessels, and instruments. The Babcock & Wilcox Company is cooperating with BNL on this project and will operate the loop.

Several loops were operated chiefly to determine the corrosiveness of steels and welds to flowing Bi. The most significant of these was Loop G, which is a large, forced circulation loop. It is capable of providing the highest linear velocities

of any corrosion loop thus far operated. In March, a 2653-hr test was completed on this loop; the results are discussed in the next section, "Corrosion and Mass Transfer." The loop is now being revised to test a duplicate of the in-pile section of Radiation Loop No. 1, described under the heading "Radiation Effects."

#### Corrosion and Mass Transfer

The thermal convection loop program has yielded new and valuable results on the corrosion behavior of steels in contact with U-Bi containing Zr and Mg inhibitors:

1) Higher hot leg temperatures do not increase the rate of corrosion of 2 $\frac{1}{4}$  Cr - 1 Mo steel. This may be due to an improvement in protective film formation with an increase in temperature.

2) A carbon steel containing Ti has been found to be exceptionally resistant to corrosion and mass transfer. ZrC films were found on the surface of tabs of this steel after contact with Bi containing Mg and Zr.

3) Pitting was observed in two 2 $\frac{1}{4}$  Cr - 1 Mo steel loops operating isothermally. Examination suggests their surfaces may have been altered upon heat treatment with argon.

4) In one loop, penetration of Bi through a 5 Cr -  $\frac{1}{2}$  Mo weld was found, accompanied by rapid oxidation of the outside of the pipe in the vicinity of the leak. This was also observed in control tests with intentional leaks. Under limited access to air (i.e., under insulating tape) Bi appears to promote catastrophic oxidation of 2 $\frac{1}{4}$  Cr - 1 Mo steel by air.

5) Gas-nitriding the inner surface of 2 $\frac{1}{4}$  Cr - 1 Mo steel increases its resistance to corrosion.

6) The low chromium steels (1 $\frac{1}{4}$  Cr -  $\frac{1}{2}$  Mo,  $\frac{1}{2}$  Cr -  $\frac{1}{2}$  Mo) and carbon steels are not detectably corroded in thermal convection tests. The high chromium steels (5 Cr -  $\frac{1}{2}$  Mo and some 2 $\frac{1}{4}$  Cr - 1 Mo) are less resistant to corrosion under the same operating conditions.

7) Tellurium is not effective as a corrosion inhibitor.

The new loops which have been put into operation are made of carbon steel and low alloy steels.

Two electromagnetically pumped corrosion test loops have operated for 6750 and 8397 hr. Slight corrosion has been detected by radiography at the 4 to 6 Cr welds of the latter loop (2 $\frac{1}{4}$  Cr - 1 Mo steel) while the former loop (1 $\frac{1}{4}$  Cr -  $\frac{1}{2}$  Mo steel) shows no apparent corrosion with the ex-

ception of welds in two thermocouple wells. A slight deposit is present on the walls of the coolers of both loops. The additive concentration of Zr, Mg, and U in each loop has stabilized at the design specification, no additions having been made since start-up. Two mechanically pumped loops capable of delivering up to 11 gpm are under construction. One loop is being constructed of low carbon steel and 2½ Cr - 1 Mo steel, and the other of 1¼ Cr - ½ Mo steel.

The best hard bearing material tested so far for service in U-Bi containing Mg and Zr is  $\text{Al}_2\text{O}_3$  flame-coated on AISI 4130 steel. Graphitar vs tool steel and Mo vs Rex AA or Stellite 90 have shown the best results of the soft vs hard combinations tested; wear on the softer component has been smooth but excessive.

Surface reactions between steels and liquid Bi alloys are being studied in order to obtain a better understanding of corrosion phenomena. The following new observations were made:

1) No steel tested has ever corroded in Bi solutions containing more than 2 ppm Zr at isothermal temperatures up to 820°C.

2) Removal of ZrN films and repeated contacting with a Zr-Bi solution at 780°C indicate that rehealing of ZrN films occurs at this temperature. After the nitrogen level in the steel is reduced by repeated film removal, ZrC layers begin to form, and the steel does not corrode.

3) The factors determining whether nitride or carbide films will form on a given steel seem to be nearly independent of the steel composition, but consistently dependent on the relative content of combined nitrogen (ester-halogen insoluble, e.g., aluminum or titanium nitrides) in the steel. Steels on which nitride films form have a small percentage of their total nitrogen combined, while steels on which carbide films form have 50 to 100 % of their total nitrogen as ester-halogen insoluble. Ti in steels promotes formation of carbide surface films.

4) A method has been developed for stripping ZrN or ZrC films from steel surfaces. The effects of steel composition, nitrogen activity, and carbon activity on corrosion and film formation can now be studied by microanalysis, electron microscopy, and electron diffraction of stripped films. An electron microscope and diffraction apparatus are currently being installed.

5) Tracer experiments have shown preferential deposition of radioactive Zr on high Cr steels and

on 5 Cr welds in 2½ Cr - 1 Mo steels at 575°C from Bi solutions. A carbon steel sample also picked up a heavy deposit. The films are too thin for identification of their structure by x-ray diffraction.

#### Fuel Stability Studies

The solubility of U in Bi has been redetermined (Figure 5). It can be represented by the equation

$$\log_{10}(\text{ppm U}) = 6.586 - (2240/T) (\pm 2\%)$$

over the temperature range 300° to 480°C, and by the equation

$$\log_{10}(\text{ppm U}) = 7.163 - (2774/T) (\pm 10\%)$$

over a temperature range 480° to 725°C. The solubilities of Te, Mn, and Ni in Bi have also been determined. The liquidus curves of the U-Zr-Bi system at 350°, 375°, 400°, and 425°C have been determined for the Bi-rich portion of the phase diagram. These are shown in Figure 6. The individual and combined effects of Zr, Mg, Na, and Li on the U and Zr solubility are being investigated. Na(1800 ppm) appears to increase the U solubility in Bi by  $\approx 25\%$ . Anomalous solubility effects have been found in graphite systems containing small excesses of U. Large amounts of U are removed from solution at temperatures below 500°C. The U can be driven back into solution at temperatures above 575°C, which indicates that the reaction is reversible.

The oxidation rates of alloying constituents from liquid Bi are being measured. The oxidation of U from U-Mg-Zr-Bi solutions was found to be catalyzed by the presence of reacted U (presumably  $\text{UO}_2$ ). The oxidation of Mg from Bi is first

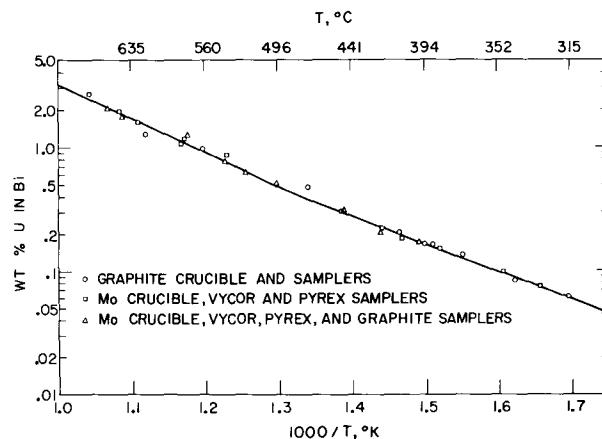
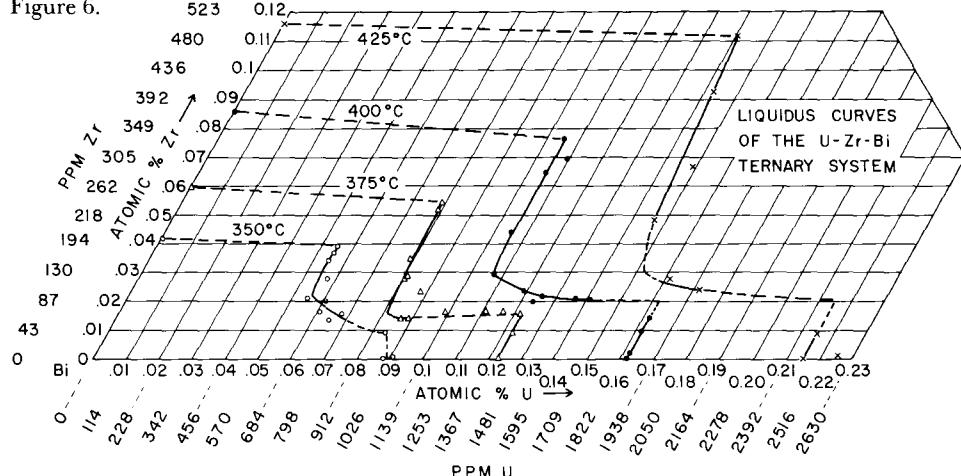


Figure 5. The solubility of uranium in bismuth.

Figure 6.



order with respect to the Mg concentration when the air pressure is held constant. The oxidation rate of Mg in Mg-U-Bi solutions is increased when reacted U is present. Mg will protect U from oxidation out of Bi in certain concentration ratios that can be predicted from one experimentally determined ratio at the same temperature by means of the equilibrium constant for the reaction  $2\text{Mg}_{(\text{Bi})} + \text{UO}_2 \rightleftharpoons 2\text{MgO} + \text{U}_{(\text{Bi})}$ , assuming the activity coefficients for U and Mg remain constant.

Mg additions to Bi have been found quantitatively to reduce uranium oxide, calcium oxide, and zirconium oxide, and to produce solutions of these metals in Bi.

#### Radiation Effects

Radiation Loop No. 1 has been under construction since the fall of 1957. This is a cooperative project between BNL and the Babcock & Wilcox Company's Atomic Energy Division. The major components, including the control panel, the out-of-pile section, and the in-pile portion, are completed. To date the thermocouples and power leads have been connected and tested between the control and out-of-pile portion. The pump, deposition gauge, and velocity gauge have been placed in position and the loop found vacuum tight. The out-of-pile run to be made this fall will consume  $\approx 1000$  hr of running time; the loop will then be placed in the reactor.

The design of a high radiation level cell in which metals may be processed and examined has been completed, and construction should be finished in the fall of 1958. The first examination will be on Radiation Loop No. 1. An intermediate

metallurgical cell has been designed and built to examine samples with an activity range of several hundred r/hr. In particular, BNL capsules containing samples submerged in U-Bi solutions are being examined for changes produced in steel and graphite. To date  $\approx 12$  capsules have been examined, and some indications of radiation-induced corrosion of steel samples have been found. The graphite does not seem to be affected by the fission recoil, although some uncertainty exists as to the stability of any layers put on graphite.

#### Graphite Studies

These studies have been undertaken to determine the feasibility of using graphite as a moderator. Transmission experiments in which Bi was passed through a given thickness of graphite showed that the macropore structure of the graphite is important up to a thickness of  $\frac{1}{8}$  in. Beyond this thickness (up to  $\frac{3}{4}$  in.), the micropore structure is the controlling factor. Between these thicknesses, the Darcy equation seems to apply. Thermal conductivity of pre- and postirradiated graphite has been determined in the temperature region 20° to 600°C (Figure 7). For LMF reactor purposes, the results indicate a reduction of 20 to 50% in thermal conductivity because of neutron bombardment. This is encouraging from an engineering point of view, and further work is under way to measure the effect of fission recoils on conductivity due to U penetration into the graphite.

#### Blanket Development

Study has continued of the possible use of a slurry of solid  $\text{ThBi}_2$  particles in liquid Bi as a fluid breeder-blanket material. X-ray diffraction studies

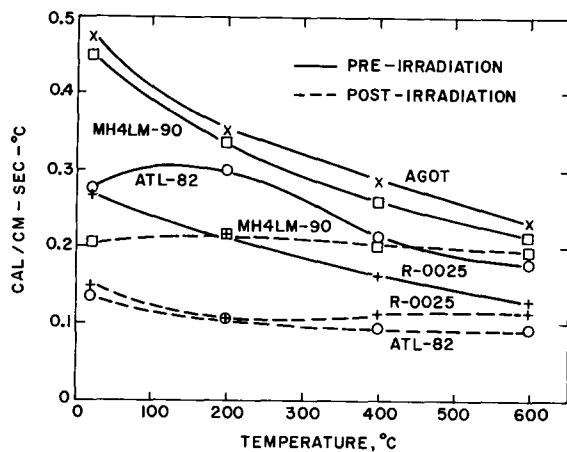


Figure 7. Thermal conductivity as a function of temperature before and after irradiation. Exposure of  $3.6 \times 10^9$  nvt at  $360^\circ$  to  $420^\circ$ C in the MTR.

have confirmed the tetragonal crystal structure and lattice dimensions of  $\text{ThBi}_2$  as determined by Ferro. The habit plane of  $\text{ThBi}_2$  platelets precipitating from solution in liquid Bi is the (001) crystal plane. The Rockwell superficial hardness of  $\text{ThBi}_2$  is  $\approx 15\text{-T-60}$ .

A suggested method for reconstituting Th-Bi slurries involves heating to dissolve the solids and then cooling rapidly to precipitate the solids as very fine platelets. Viscous dispersions of  $\text{ThBi}_2$  platelets 50  $\mu$  or less in diameter can be rendered fluid in concentrations up to 10 wt % Th by heating at  $800^\circ$ C for 20 min or at  $900^\circ$ C for 5 min. These heat treatments decrease the particle diameter-to-thickness ratio from 50:1 to 5:1. American Metal Climax Inc., under subcontract to BNL, has shown that large-scale production of fine platelet dispersions can be accomplished by gas atomization of Th-Bi solutions. Aeroprojects Inc. has demonstrated that application of ultrasonic energy to a Th-Bi alloy during cooling from a temperature above the liquidus causes formation of small, relatively equiaxed  $\text{ThBi}_2$  particles rather than platelets. The company is investigating this method further.

Th-Bi slurries cycled between  $350^\circ$  and  $580^\circ$ C at 6 cy/min for 500 hr in graphite tubes have exhibited accelerated particle growth, primarily by particle agglomeration, but no deposition of  $\text{ThBi}_2$  on the tube walls in the cooler end.  $\text{ThBi}_2$  was completely deposited as large columnar grains in the cold ends of similar Croloy 2 $\frac{1}{4}$  tubes. A 10

wt % Th slurry containing 0.10 wt % Te showed no particle growth, agglomeration, or deposition after this treatment in either graphite or steel tubes. Addition of 0.05 wt % Te to a 5 wt % Th-Bi slurry also prevented pressure-welding and particle agglomeration in the slurry during 500 hr of tumbling at  $525^\circ$ C.

Te additions to Th-Bi slurries in the ratio of one part by weight Te to 100 parts Th prevent the growth of  $60\text{-}\mu$   $\text{ThBi}_2$  particles under conditions which cause particle growth to  $120\text{ }\mu$  in slurries without Te. Te additions in this ratio slow down, but do not prevent, the growth of smaller particles and the thickening of fine  $\text{ThBi}_2$  platelets. Dispersions containing Te are best prepared by heating Th chips or powder in molten Te-Bi alloys. The time required for complete reaction is longer than that for Th-Bi slurries without Te. Addition of Te to a Th-Bi slurry causes the originally dense single crystals of  $\text{ThBi}_2$  to become hollow. The mechanism by which Te additions inhibit  $\text{ThBi}_2$  particle growth, agglomeration, and deposition during thermal cycling is not yet understood. At  $1000^\circ$ C, Te lowers the solubility of Th in Bi markedly, but at lower temperatures the Te appears to be concentrated almost completely in the solid phase, and has little detectable effect upon the Th solubility. X-ray diffraction studies of  $\text{ThBi}_2$  containing Te have failed to reveal any effect upon the crystal structure or lattice dimensions.

During the past year, a third pumped slurry loop was constructed in an effort to learn more about the properties of thorium bismuthide slurries and to study the deposition of  $\text{ThBi}_2$  on cold walls. Results obtained from it show that  $\text{ThBi}_2$  slurries can be circulated isothermally for long periods at velocities from 0.4 to 2.0 ft/sec without difficulty. However, the circulation of these slurries through even a small temperature differential ( $25^\circ$ C bulk) results in the precipitation of  $\text{ThBi}_2$  unless Te has been added to the slurry in amounts of about Th:Te = 10:1 (atomic ratio).

A program has been started to investigate dispersions of insoluble Th compounds such as the oxide, carbide, and fluoride in liquid Bi as possible fluid breeder-blanket materials. Thorium oxide has a density very close to that of liquid Bi and should form dispersions that are stable with respect to particle settling. Fluid dispersions containing up to 10 wt %  $\text{ThO}_2$  have been formed by using Mg and Zr additives to promote melting. Both elements are partly removed from solution

in Bi during dispersion formation. Factors tending to increase the amount of  $\text{ThO}_2$  that can be dispersed include increasing the additive content and  $\text{ThO}_2$  particle size; increasing the time, violence, and temperature of stirring; and stirring under vacuum rather than inert gas. In dispersions containing 10 wt %  $\text{ThO}_2$ , increasing the additive content increases the uniformity of particle distribution.

Slurries of  $\text{ThO}_2$  containing  $\text{UO}_2$  in solid solution dispersed in low melting, low cross section liquid metals might be used as a combination of fuel and fertile material in a one-region thermal reactor.  $\text{ThO}_2\text{-UO}_2$  solid-solution powders containing approximately 1.5 wt % U and 5 wt % U have been dispersed in amounts up to 10 % by weight of powder in liquid Bi and Pb-Bi eutectic, with Mg as a wetting agent.

## GENERAL REACTOR PHYSICS

### Experimental

Some additional data have been accumulated during the past year in the basic program of measuring lattice parameters of water moderated, slightly enriched uranium rod fueled reactor cores. Measurements on the rod lattices in the 0.387-in.-diameter size have virtually been completed. Some of the intensive parameters in these lattices were remeasured during the year. Studies of the 0.250-in.-diameter rod lattices have comprised the chief effort. Since the 1.3% enriched rods are being retained in the 0.387-in. size by the Westinghouse Electric Corporation, the 0.250-in. lattices will be limited to the 1.0% and 1.15% enrichments. A paper comprising all the results to date was prepared for the second Geneva Conference. Much effort has been devoted to improving techniques of measurement of lattice parameters in assemblies of small rods.

The slightly enriched uranium plates for the plate lattice program were received from the vendor. A number of these elements had to be returned for reworking, and the plate lattice program has therefore been delayed until next year.

A request for uranium oxide fuel for critical experiments was initiated.

Measurements were performed on the activation distribution in large (3.6-in.) diameter natural uranium rods. The results show that the same distribution is obtained with fission catcher foils

and with dysprosium, and justify the use of bare dysprosium foils for intracell flux measurements under very severe conditions.

An approach to critical was made with the new, enriched, graphite reactor fuel elements in water to establish specifications for safe storage in the canal.

### Theoretical

Theoretical studies have been continued on resonance escape probability, on analyses of the proposed high flux research reactor, and on the characteristics of the reloaded graphite reactor. Basic contributions have been made to the theory of neutron thermalization. Miscellaneous work has included investigation of LMFR problems concerned with stability and hazards, transport theory studies of neutron leakage from hydrogenous systems, medium interaction perturbation theory studies, and applications of irreversible statistical mechanics to neutron spectrum problems.

The purpose of work on resonance escape probability of neutrons with respect to  $\text{U}^{238}$  and  $\text{Th}^{232}$  has been to obtain a fundamental understanding of physical phenomena and methods of calculation. Attention has been concentrated on hydrogen moderated systems, although many of the results are more general. Some cases have been studied, and results obtained, by Monte Carlo procedure on a digital computer with all physical effects included. This has been supplemented by analytical methods that often include simplifications.

A fruitful analytical development has been the extension of the Wigner resonance integral concept to the treatment of lattices. Thus lattices, lumps of absorber, and homogeneous systems are treated in a unified manner. For lattices the interaction effects of neighboring absorbers, or so-called Dancoff corrections, occur as an inherent part of the theory. The differing geometric forms are related semiquantitatively by equivalence theorems. This approach leads to practical calculations of absorptions in a particular resonance by a method of determinable accuracy, and the method is applicable to a wide class of assemblies of practical interest. A result of these studies is that interference between resonance and potential scattering as well as Doppler broadening, and the interaction between these two effects, are quantitatively important.

Monte Carlo calculations of resonance escape probability on the Univac have been completed for a number of cases. These consider the absorptions in 18 resolved resonances of  $U^{238}$  in water moderated rod lattices, for which careful measurements exist. The results give insight into several physical effects. Energy degradation by  $U^{238}$  is important in individual resonances, but its net effect tends to cancel out in summing over resonances lying below several hundred ev in energy. Therefore, the omission of degradation effects, which greatly simplifies the mathematics, appears justified for low lying resonances. The shape of the spatial distribution of absorptions is remarkably similar for the various low lying resonances.

Some work has been carried out with the Applied Mathematics Division on the development of a digital computation procedure for the IBM 704 to solve the slowing-down integral equations and resonance capture problem for homogeneous mixtures. This procedure is faster than the Monte Carlo method. By the use of equivalence theorems, some results for homogeneous systems can be extended to heterogeneous systems.

The influence of a higher resonance upon captures in a resonance lying at a lower energy is being investigated by a variational method for the case of a homogeneous mixture.

A paper prepared jointly with the Argonne National Laboratory for the second Geneva Conference describes some of the research on resonance escape probability.

Calculations of characteristics of the proposed high flux research reactor have been continued by multigroup methods, and a paper describing some of the results has been prepared for the 1958 Geneva meeting. Comparison of calculations with Olcott's experiments on similar  $D_2O$  moderated assemblies shows that a noticeable improvement in predicted critical masses occurs if the effect of chemical binding on the low energy neutron spectrum is included. Cases have been calculated to confirm the general design concept and to indicate characteristics which should be studied further. It has been ascertained that neutron fluxes in the intermediate and the thermal energy ranges can be obtained in the core and reflector respectively, each of which has an intensity between  $10^{14}$  and  $10^{15}$  neutrons/cm<sup>2</sup>-sec. A high fuel concentration in the alloy is shown to sacrifice epithermal flux to some extent for fixed power per unit vol-

ume and to result in smaller cores having several advantages. Further investigation of the effects of distribution of fuel concentration and of questions related to xenon, temperature, and burn-out effects are to be studied by a more appropriate multigroup coding, the GNU.

Calculations have been concluded on several aspects of the reloading of the BNL graphite reactor with enriched fuel elements. To guide the experimental work, critical sizes, flux distributions, and xenon effects have been predicted for several different geometric forms of a core reloaded entirely with enriched fuel. It was shown that a loading having an elongated rather than circular cross section over faces from which neutron beams are extracted should increase the beam intensity available at the outermost beam holes by a factor of 3 for 35% increase in critical mass and 20% increase in pumping power. A paper describing some aspects of these reloading studies has been submitted to the second Geneva Conference.

An analytical solution of the neutron thermalization problems has been developed which is applicable to homogeneous mixtures having a quite general form of the kernel, and which describes energy exchange between neutron and moderator atoms. This method facilitates investigation of the accuracy of various approximate models used previously and permits improved calculations to be made where required. For deuterium moderator and heavy absorption a significant correction has been noted to the conventional first moment expansion.

In collaboration with the Physics Department, studies have been continued on the level structure and inelastic scattering properties of intermediate and heavy nuclei.

## FUEL PROCESSING (GENERAL)

### Fluoride Volatility Project

For several years, the fluoride volatility program dealt with the use of interhalogen mixtures consisting mostly of  $BrF_3$  for dissolving natural uranium fuel elements, prior to recovery and decontamination of the U as  $UF_6$  by distillation. On May 15, 1957, a pilot plant for the dissolution process suffered major damage when a series of explosions occurred in rapid succession during a run. The pilot plant was later dismantled and put in storage, pending a decision from the Atomic

Energy Commission as to whether or not it would be re-erected in a special building of its own.

Since the cause of the accident could not be definitely determined, it was decided to carry out a program of experiments designed to give the threshold conditions under which an explosive reaction can occur between natural U and certain interhalogen mixtures. A test facility was built, and several runs have been made, but in only one run was the reaction rate comparable to that of an explosion. The conditions for this run were not significantly different from those for the preceding ones, the U being exposed at 171°C to vapor produced from a solution consisting of 1.2 M %  $\text{UF}_6$ , 1.2 M %  $\text{Br}_2$ , and 97.6 M %  $\text{BrF}_3$ . At the instant of explosion the U temperature was 148°C. Attempts will be made to obtain further explosive reactions.

#### **Reprocessing of Power Reactor Fuels**

Exploratory studies have been undertaken of the problems involved in the recovery of U and Pu from the Enrico Fermi reactor fuel. This fuel, Zr-jacketed 10% Mo - 90% U alloy, may be prepared for solvent extraction by dissolution in mixed nitric and hydrofluoric acids. In the proposed process both the jacket and the core are dissolved in one operation in 2.5 M  $\text{HNO}_3$ -0.06 M HF. The core dissolves at a rate of about 100 mg/cm<sup>2</sup>-min. At U concentrations up to 12 g/liter, Mo is soluble. Varying the  $\text{HNO}_3$  concentration over the range 2.5 to 5.5 M at a constant HF molarity of 0.06 and a F/Zr mole ratio of 9.87 had no effect on the time required for complete dissolution,  $\approx$ 4.7 hr. When the HF concentration was reduced to 0.04 M and the F/Zr ratio to 6.57, the total dissolution time was 3 hr for 5.5 M  $\text{HNO}_3$  and 8 hr for 2.5 M  $\text{HNO}_3$ . A precipitate formed occasionally when the  $\text{HNO}_3$  concentration was less than 4.5 M. The precipitate was dissolved by adding an oxidizing agent such as  $\text{CrO}_3$  to the light slurry and refluxing for an additional half hour, or simply by refluxing for about 10 hr. About 7 moles  $\text{CrO}_3$  were required per mole U in the precipitate. Development of a reprocessing method for Enrico Fermi reactor fuel has been confined to laboratory-scale studies with unirradiated fuel samples.

#### **Fuel Element Reprocessing in Gas-Fluidized Beds**

In the reprocessing of spent solid fuels from nuclear reactors by volatilization processes, one of the major problems is the removal of heat from

the highly exothermic reactions between the metallic element and the reactant gas. The problem is compounded by the elevated temperatures required, the high reaction rates, and the low heat capacities of the gaseous effluents. The use of a liquid carrier (fused salts, interhalogens, etc.) for the gaseous reactant is an obvious means of removing the heat, and this principle is utilized in a number of well-known fuel process developments.

Recent work at BNL has demonstrated, however, that a gas-fluidized bed of granular solids, by virtue of its high capacity for heat transfer, offers a very desirable set of advantages in controlling such reactions. Among these are: 1) that the rapid removal of the reaction products brings about large increases in the surface reaction rates and the uniformity of the reaction; 2) that the granular material may be selected so as to be completely inert in the system; and 3) that the heat removal capacity of gas-fluidized beds is much higher than the capacity of the gas alone and actually approaches that of a liquid.

### **FUEL DEVELOPMENT STUDY**

#### **Fluidized Solids Fuel Element**

An investigation is currently being made of the feasibility of using a fluidized bed, an assemblage of granular solids suspended in upward-flowing liquid, as a mobile fuel for a nuclear reactor. In order to meet the requirement of uniform and stabilized distribution of the fuel particles, the design calls for a system of tubular elements containing the fuel suspension ( $\text{UO}_2$  particles in liquid Na). Since the fuel suspension in such a system would behave much like a heavy liquid, the charging of fresh fuel to the reactor and discharging of spent fuel from it could be done by the adjustment of pressures in the system.

The investigation thus far involves the nuclear physics aspect of the problem and an experimental study of a simulated fuel system composed of glass beads (representing  $\text{UO}_2$ ) in water (representing Na) contained in a full-scale fuel element assembly constructed mainly of Lucite.

The experimental results clearly demonstrate that the design concept is sound from the mechanical standpoint in that the fuel suspension is entirely stable and uniform, and the granular fuel in the form of a fluidized bed can be easily charged into and discharged from the reactor.

## GRAPHITE STUDIES

The monitoring program dealing with radiation damage to graphite in the Brookhaven reactor has revealed that a large amount of growth has taken place recently. This is attributed to the change from natural uranium fuel elements to the enriched elements. Following the Windscale incident, an investigation of the burning characteristics of graphite was initiated. Tests have shown that self-sustained burning can be attained if the graphite reaches 900°C. It is a function of the air velocity at this temperature, but is independent of whether or not the graphite is irradiated. Above 1000°C, burning is independent of air velocity, and test fires had to be extinguished by suffocation. Samples of graphite from the Brookhaven and Oak Ridge reactors were annealed at 100°C. Following this treatment, stored energy continued to be released over the temperature range of 300° to 800°C. This is the first time such an effect has been measured on samples from these reactors.

## WASTE PROCESSING STUDIES

### Clay Adsorption

The continuing study of ultimate disposal of high level radioactive wastes by adsorption on columns of montmorillonite clay has led to the need for more exact information on the capacity of the clay to effect the very high decontamination of the waste streams required in full-scale operations. Accordingly, a test run was made in which a simulated waste solution, with 0.02 *N* NaNO<sub>3</sub> and spiked with 2 curies Sr<sup>90</sup> in 44 liters of solution, was passed through a system of three 5-ft columns (0.75 in. in diameter) connected in series. At the end of 12 days of operation 28 liters of the solution had passed through the system, an amount equal to about 200 column volumes, and the results showed that the decontamination factor for Sr was then  $1.6 \times 10^8$ . Figure 8 shows an autoradiograph of the first column with evidence that the radioactivity was virtually confined to the first 20 in.

### Calcination

In the study of calcination of wastes containing bulk salts of Zr and Al, to produce the stable oxides, emphasis has centered on the design and testing of equipment to insure against the escape of radioactive dust from the system, and on the

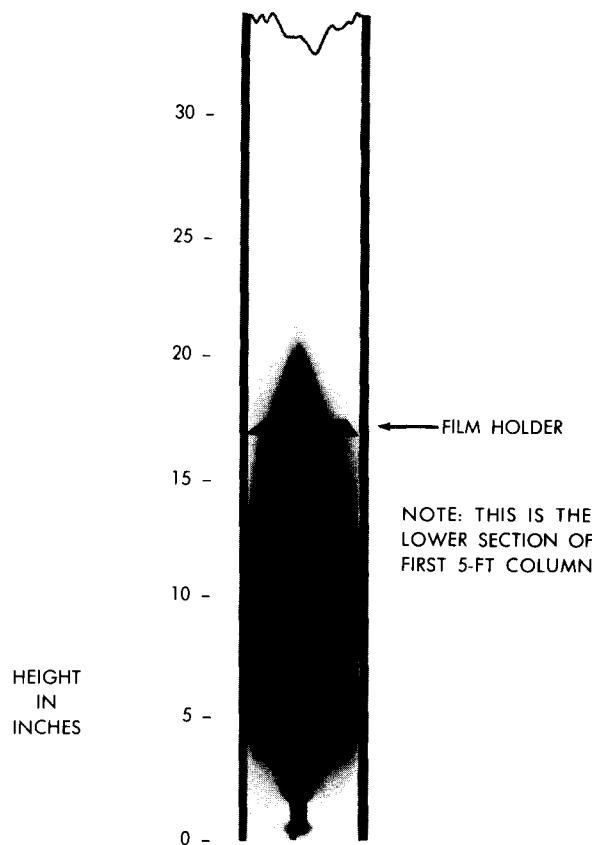


Figure 8. Autoradiograph of long clay column showing area in which  $\approx 1.2$  curies of Sr<sup>90</sup> concentrated.

determination of suitable materials of construction for calciners handling Zr wastes in which the off-gases are chiefly hydrogen fluoride, oxides of nitrogen, and steam at temperatures up to 700°C. The problem of dust control is being studied with two basic objectives in mind: 1) the selection of a type of calciner that will minimize dust production at the source, and 2) the elimination of all noncondensable gases from the system to the fullest possible extent. In line with these objectives, a 6-in.-diameter rotary kiln calciner (Bartlett Snow) connected directly to a condenser has been installed and a few test runs made with simulated aluminum nitrate wastes. Results of these tests indicate that the volume flow of noncondensable off-gases is very small and that under these conditions the condenser is a very effective trap for the entrained dust.

### Sulfex-F Dissolution

The presence of both fluorides and nitrates in the Zr waste solutions presents a potentially seri-

ous corrosion problem during calcination to the oxides. If the fluoride ion could be eliminated or reduced to an insignificant amount, the problem of waste processing for disposal would be greatly simplified. In line with the interest in the use of electrochemical methods in connection with membrane systems, anodic dissolution of Zr in aqueous systems was investigated. These studies led indirectly to the discovery that a mixture of hot ( $160^{\circ}\text{C}$ )  $\text{H}_2\text{SO}_4$  in the range of 60 to 70 wt % and HF of very low concentration ( $0.08\text{ M}$ ) would dissolve Zr alloy fuel elements at rates adequate for fuel reprocessing. Furthermore, although HF appears to be the aggressive agent, it is not consumed in the process, since the  $\text{H}_2\text{SO}_4$ , at this temperature breaks down the zirconium fluoride complex and releases HF for further attack. In this system, then, the HF acts as a catalyst. Final solutions contain only 0.0125 of the stoichiometric amount of fluoride necessary to dissolve Zr in HF alone.

An extensive series of test runs was made to show the effect of such variables as acid concentration, temperature, etc. on the rate of dissolution. In addition, a corrosion study was made on certain likely materials of construction for the dissolver vessel.

#### **Removal and Recovery of Fission Product Noble Gases**

Experimental work was continued on the development of a continuous absorption-stripping process for the removal and recovery of fission product noble gases from dissolver off-gases. The solubilities of Xe and Kr in a number of organic solvents were measured, and a correlation with the cohesive energy densities of the solvents was made. The data seemed to conform to Hildebrand's solubility theory. In addition, the solubilities of  $\text{N}_2$ ,  $\text{O}_2$ , and  $\text{N}_2\text{O}$  were measured, and single-stage separation factors were obtained. Of the organic solvents investigated, those with a kerosene base (e.g., Amsco 123-15) had the most desirable characteristics as process absorbents. The solubility increased with decreasing temperature.

It was recognized that the properties of  $\text{N}_2\text{O}$  were too similar to those of Xe to allow the separation of these two gases in an absorption-stripping operation. For this reason it was predicted that  $\text{N}_2\text{O}$  would be an excellent solvent for Xe and Kr. By designing the absorption-stripping process for low temperature and elevated pressure

operation, the bulk of the  $\text{N}_2\text{O}$  could easily be removed from the dissolver off-gas stream. Measurement confirmed the prediction. The solubilities of Xe and Kr in  $\text{N}_2\text{O}$  at  $-80^{\circ}\text{C}$  were found to be 60.5 and 8.7 cc (STP) per cc-atmos respectively, and a single-stage operation factor was obtained in a nitrogen carrier stream. A plant design was completed using  $\text{N}_2\text{O}$  as process solvent and the absorption-fractionation principle to obtain a high concentration of noble gases in a two-column operation.

Since  $\text{N}_2\text{O}$  is an endothermic compound, its use might present a hazard. The Freons possess chemical properties which would provide for stable operation. Solubility measurements were made and showed surprisingly high absorbency for Xe and Kr, 55.3 and 12.6 cc (STP) per cc-atmos respectively, with a single-stage separation factor of 17.5 for separation of Kr from  $\text{N}_2$ . A plant design using Freon-12 as process fluid is now being drawn up.

Data have also been obtained on the adsorption capacity of Xe and Kr on molecular sieves.

#### **GAMMA RADIATION STUDIES**

To supplement information for the engineering design of the High Intensity Food Irradiator (HIFI) to be built for the Quartermaster Corps, BNL has been requested to obtain data by using low level  $\text{Co}^{60}$  sources in various slab arrangements. The prime objective of the BNL work is to examine experimentally QMC specifications for the HIFI and to determine whether or not  $\text{Co}^{60}$  can be used as the irradiator source material.

Depth-dose measurements were made in simulated food packages by using various arrangements of source material. The relation between the activities required and the dose rate at any point was obtained. Results were extrapolated to a full-scale system, and the required total and specific activities determined. Specifications could be met by using two parallel irradiator slabs  $\approx 10$  ft long and  $2\frac{1}{2}$  ft wide, containing a total of  $\approx 3.8 \times 10^6$  curies with an average specific activity of 67 C/g. Further experimental work with  $\text{Co}^{60}$  will be undertaken during the 1959 fiscal period.

A comparative study was made of  $\text{Co}^{60}$ ,  $\text{Cs}^{137}$ , and  $\text{Na}^{24}$  as potential irradiation sources. The HIFI specifications were used as a basis of comparison. Hand calculation techniques were used to determine depth-dose relationship in an infinite

slab system for material of unit density. Data obtained from measurements of gamma dose distribution in paraffin from rectangular  $\text{Co}^{60}$  slabs were then used to correct these calculations for the finite system considered. The general conclusions were as follows:  $\text{Co}^{60}$  and  $\text{Na}^{24}$  could meet the specifications, but the specific activity required for  $\text{Na}^{24}$  would be greater than that produced in any sodium-cooled reactor built or to be built in the near future. The  $\text{Cs}^{137}$  did not quite meet the maximum-to-minimum dose requirement of 1.2 in the systems considered in this study. Further, the specific activity required and the total curies required were not available. Experimental work will be undertaken, with both  $\text{Na}^{24}$  and  $\text{Cs}^{137}$  used as irradiation sources, to check the results of this study.

### Graft Copolymerization

The results on grafting of styrene to preirradiated polyethylene have been summarized in a paper for the International Polymer Conference in Nottingham, England, in July 1958.

Attempts to graft gaseous monomers like butadiene and trifluoromonochloroethylene to preirradiated polyethylene were unsuccessful. Success was achieved, however, in the case of butadiene liquid monomer and butadiene-styrene mixtures.

An investigation has begun on the detection and estimation of free radical populations in irradiated polyethylene by use of magnetic resonance techniques.

### Solid State Polymerization

A restudy of the solid state polymerization of acrylamide and other crystalline monomers indicates that appreciable postirradiation reaction occurs over periods of many months. Photomicrographic studies show that the reaction is nonhomogeneous and that noncrystalline polymers are formed.

### Radiation Catalysis

It has been found that irradiation of reduced molybdenum oxide, alumina-molybdena, and gamma alumina in the presence of hydrogen increased the ability of these oxides to convert *ortho*-*para* hydrogen mixtures. A true catalytic effect is believed to be operating, since positive results are obtained at  $-78^{\circ}\text{C}$  where a paramagnetic effect is not usually found. The postirradiation retention of catalytic activity is greatly affected by tempera-

ture, atmospheric gases, and vacuum. At present the effect appears to be due to a weak catalyst-hydrogen complex.

### HEAT TRANSFER STUDIES

A mercury heat transfer research program has been conducted by the Nuclear Engineering Department for several years. The objective of the work is to determine heat transfer coefficients for mercury flowing on the shell side of staggered tube banks and to correlate the results with the important variables. The cross-flow portion of the program is completed, and during the past year the first in-line flow results have been obtained. A certain lack of reproducibility of results has slowed progress in recent months, but it is hoped that revisions now being made in the apparatus will remedy the situation. In Figure 9, results are shown for chrome-plated unwetted tubes. Both the Nusselt and Peclet numbers are based on the conventional hydraulic radius principle. The results appear quite reasonable, in view of the fact that the Nusselt number for slug flow for in-line flow outside of tubes is roughly twice that for the same kind of flow inside tubes.

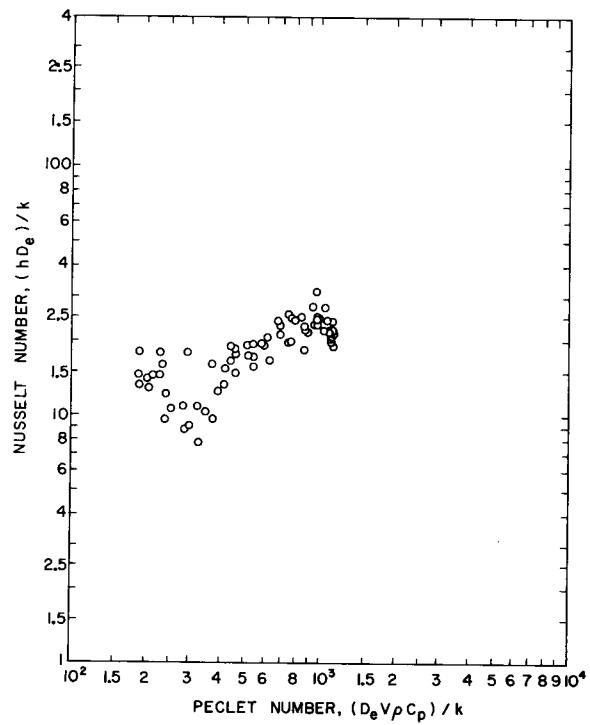


Figure 9. Heat transfer rates to mercury flowing parallel to and outside of tubes in an equilaterally staggered tube bank.

## SPECIAL RADIOISOTOPE DEVELOPMENT

### Yttrium-90

Long-term tests have continued on the system devised for separating the important beta-emitting isotope  $Y^{90}$  from its long-lived beta-emitting Sr parent. These tests, in progress for two years, employ a simple and convenient device, a Dowex-50 ion exchange column on which 28-year  $Sr^{90}$  is firmly fixed, to produce a  $Y^{90}$  product with a satisfactorily low  $Sr^{90}$  contamination. A simple, rapid, and eminently satisfactory method of assaying for  $Sr^{90}$  contamination in the milked  $Y^{90}$  product *before* administration to patients has been developed. Some work was started in order to determine the feasibility of using for the  $Y^{90}$ - $Sr^{90}$  separation an alumina column, which would be relatively resistant to radiation damage and which should be capable of handling several curies of activity. The mechanism of bonding  $Y^{90}$  to an anion exchange column in the hydroxyl form has been found to be via a citrate complex rather than as precipitated yttrium hydroxide. A prototype  $Y^{90}$  generator containing 113 mC  $Sr^{90}$  was delivered to one of the New York City hospitals for field testing and clinical evaluation. Several high activity  $Y^{90}$  sources have been prepared for others doing experiments in connection with the parity principle, while several smaller sources have been supplied to the BNL Medical Department in connection with a study on antibody production in mice.

### Magnesium-28

A new isolation process was developed which gives yields of  $\approx 78\%$  and product purities of  $\approx 99.95\%$  and which will permit scale-up of the present  $Mg^{28}$  production process.

### Fluorine-18

A new isolation and purification process, which reduces the handling required and is capable of being scaled up, was developed for the production of  $F^{18}$ .

### Iodine-132

The new type of  $I^{132}$  generator which utilizes the sorption of parent  $Te^{132}$  on alumina was further studied, and much was learned about the behavior of iodine, iodate, elemental iodine, and

Te on alumina. These new generators are now in routine production and are eminently satisfactory.

A new process for the production of  $Te^{132}$  has been developed, the equipment constructed, and the process put into actual routine use. This process, which is simpler and more rapid than the previous one, uses the sorption of  $Te^{132}$  on alumina as a means of separating this isotope from U and mixed fission products. The sources now used for the production of  $Te^{132}$  are surplus or rejected sections of the Al-enriched U alloy used for the new fuel elements in the reloaded BNL reactor. Te product yields are  $\approx 83\%$ , and the radiocontamination amounts to  $<0.1\%$ .

### Iodine-133

The  $Te^{130}(\alpha, p)I^{133}$  reaction in the BNL 60-in. cyclotron was studied as a possible source of  $I^{133}$  free of both stable carrier and  $I^{131}$ . The best results actually obtained gave an  $I^{131}/I^{133}$  ratio of 0.03, but extrapolation from the data obtained so far indicates that a ratio of 0.002 should be possible. While this represents a distinct improvement over the best possible ratio obtainable from fission products, where the minimum  $I^{131}/I^{133}$  ratio is no lower than 0.2, the production of therapeutic quantities of  $I^{133}$  by this process does not appear to be economically feasible. The cyclotron method may be useful in special work for which isotopically pure  $I^{133}$  would be worth the premium price.

### Samarium-151

A method of producing pure  $Sm^{151}$  from U and a fission product solution is being developed. The method will involve the use of Dowex-1 and Dowex-50 ion exchange columns coupled with ammoniacal precipitation of the rare earths group with Sr used as hold-back carrier.

### Oxygen-20

A search for the hitherto undiscovered isotope  $O^{20}$  via the  $O^{18}(t, p)O^{20}$  reaction was made by irradiating several samples of lithium hydroxide enriched to 96%  $Li^6$  and 89%  $O^{18}$  in the Brookhaven research reactor, the  $O^{18}$  being bombarded by tritons produced by the familiar  $Li^6(n, \alpha)H^3$  reaction. No evidence for the existence of  $O^{20}$  was found. As a result of this work an upper limit of  $\approx 30$  sec seems probable for the half-life of  $O^{20}$ .

### Arsenic-72

In an effort to provide an adequate and economical supply of the important positron-emitting isotope  $\text{As}^{72}$ , which is useful in locating brain tumors, a study was undertaken of the possibility of producing 9-day  $\text{Se}^{72}$  by the  $\text{Ge}^{70}(\alpha, 2n)\text{Se}^{72}$  reaction and milking the 26-hr  $\text{As}^{72}$  daughter from the Se. An analytical method was developed for separating As from Se prior to counting. Two cyclotron runs were performed in which  $\text{Se}^{72}$  was produced. A tentative milking system for separating the  $\text{As}^{72}$  routinely from the  $\text{Se}^{72}$  has been devised, but difficulties are being experienced in correlating the behavior of the  $\text{As}^{72}$  with that of the  $\text{As}^{76}$  used as a tracer.

### Molybdenum-99

A method involving sorption on alumina was developed for separating carrier-free  $\text{Mo}^{99}$  from fission product solution. The product is virtually free of other contaminants. Thus this isotope, the parent of 6-hr  $\text{Tc}^{99m}$ , can be made as a by-product of the production of  $\text{Te}^{132}$ , both the  $\text{Mo}^{99}$  and  $\text{Te}^{132}$  being obtained from the same run without interference of one process with the other.

### Technetium-99m

An alumina generator has been developed from which  $\text{Te}^{99m}$  (6-hr half-life) can be milked from its 67-hr  $\text{Mo}^{99}$  parent in a process very similar to

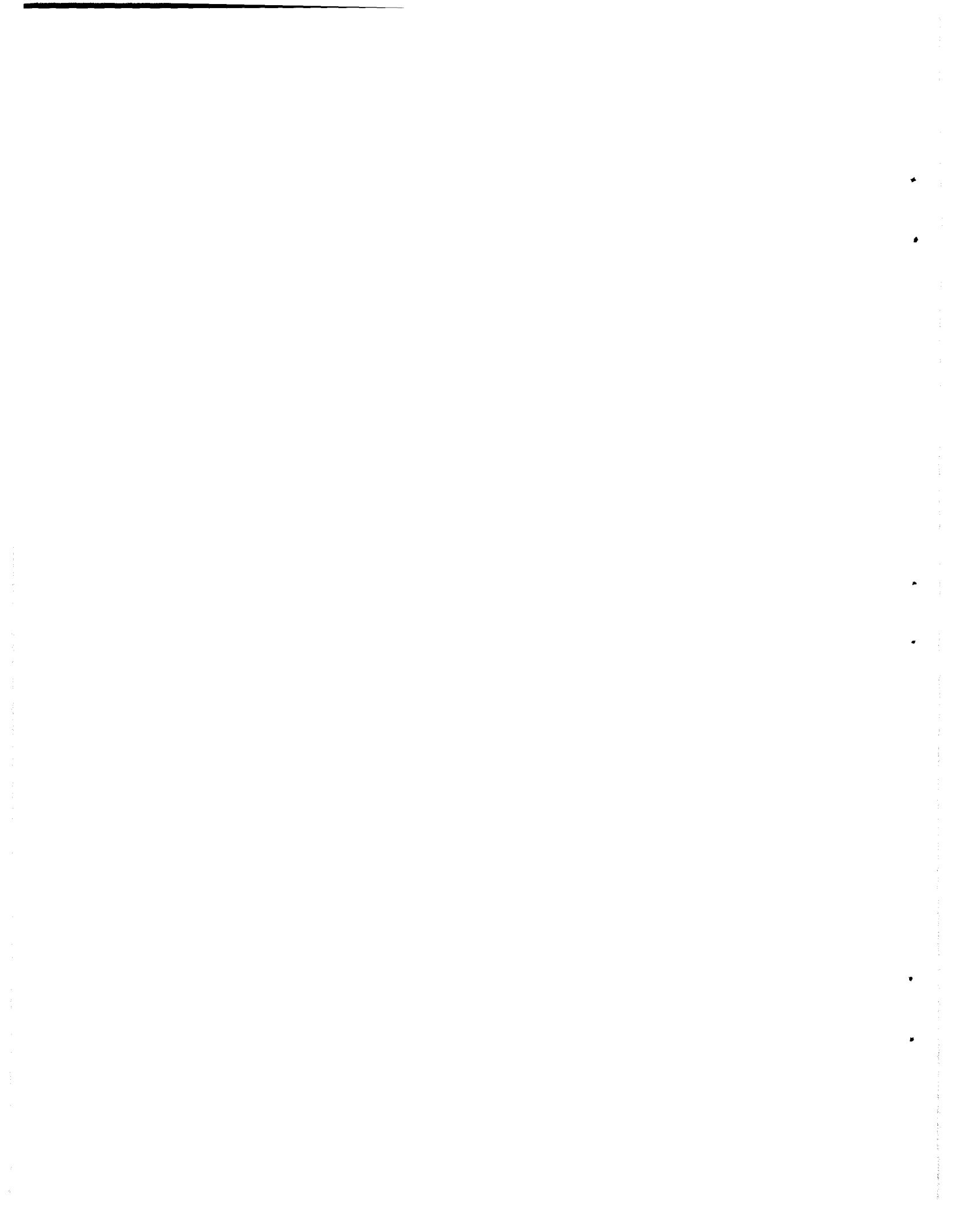
that used at present for milking  $\text{I}^{132}$  from its parent  $\text{Te}^{132}$ . This potentially useful short-lived isotope has a single gamma of 0.140-Mev energy decaying eventually to stable ( $2.1 \times 10^5$ -year half-life)  $\text{Te}^{99}$ . Te yields range between 60 and 70%, and long-lived contamination is  $\approx 0.001\%$ .

### Calcium-47

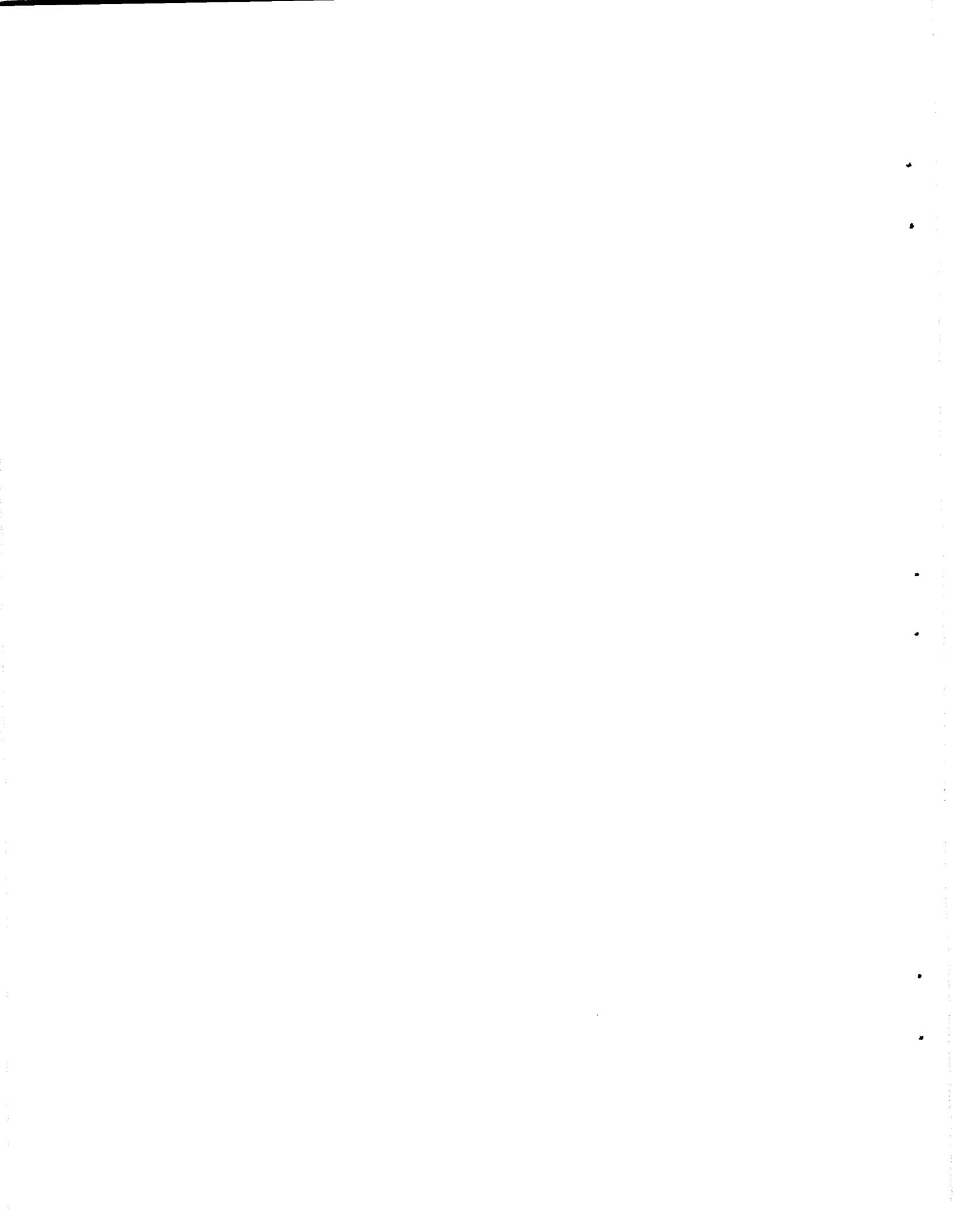
Since considerable interest in a short half-life isotope of Ca seems to exist, an investigation was begun of the possibility of producing isotopically pure 4.7-day  $\text{Ca}^{47}$  by the only method which appears to be feasible:  $\text{Ti}^{50}(n, \alpha)\text{Ca}^{47}$  in the reactor. Some work has been done on the chemical separation of the product Ca from the Ti, the process being complicated by the fact that enriched  $\text{Ti}^{50}$  would have to be used routinely and, because of its expense and rarity, would have to be recovered completely for re-use.

### Cross Section Measurements

Work has been undertaken which will lead to a measurement of the cross section for the  $\text{O}^{16}(t, n)\text{F}^{18}$  reaction as carried out in the BNL research reactor by using tritons generated by the  $\text{Li}^6(n, t)\text{He}^4$  reaction. When this is completed it will be possible to use the  $\text{O}^{16}\text{-F}^{18}$  reaction as a standard against which to measure cross sections of other similar reactions, e.g., the  $\text{Mg}^{26}(t, p)\text{Mg}^{28}$  reaction.



LIFE  
SCIENCES



## Biology

The research activities of the Biology Department continue to center on the special facilities of the Laboratory. They include, in general, studies of the effects of various kinds of radiation on biological material and the use of these studies as well as those with radioactive tracers in the elucidation of life processes. The facilities of the Department have been used to capacity, and one new section has been added to the overcrowded greenhouses. In addition, two plant growth cabinets similar to the one described in the previous annual report have been completed. The new cabinets, designed here, have better controls than the first one, which was purchased commercially.

A small section of the animal quarters is being remodeled so that mice and rats may be kept under relatively "germ-free" conditions. An important part of the Department's research program is concerned with the problem of radiation-induced aging, which involves keeping rodents over their entire life span. Minor outbreaks of disease in the present colony have been numerous enough to impede the program seriously and to make it apparent that rather drastic steps must be taken to improve the conditions under which the animals are kept. The present alterations are considered emergency measures only, to be used until adequate animal quarters can be designed and constructed.

As in the past, the influx of summer visitors has saturated the Department's facilities. In addition to 5 foreign visitors, 12 scientists and 17 students are working here this summer. This program has become an important part of the research and educational activities of the Department. During the year 41 scientists, of whom 14 were from other countries, worked in biology without compensation from the Laboratory; they contributed to the research program and at the same time gained experience in special techniques. They were here for periods ranging from a few weeks to 12 months and accounted for an appreciable fraction of the effort of the Department. In addition, many foreign and domestic scientists visited the Department for periods of a week or less.

This report will briefly outline the major areas of research in the Department and indicate the progress in each during the past year.

### MAMMALIAN PHYSIOLOGY

#### Radiation-Induced Aging

If an animal receives a large dose of radiation it may sicken and die within a month. If it recovers, it will have a decreased life expectancy, due in part to the induction of cancer, which does not appear until long after the irradiation, but seemingly also due to a general acceleration of the aging process. For this reason the phenomenon has come to be known as radiation-induced aging. If it can be proved that radiation does indeed accelerate natural aging, then it will become a powerful tool for study of the aging process. The present program is designed not only to elucidate the basic mechanism of radiation-induced aging, but to relate it to the phenomenon of natural aging. Since, even with short-lived animals such as mice, most experiments in this field take nearly two years to complete, progress often seems quite slow.

First, it was shown that shielding a very small portion of the bone marrow does not affect the course of radiation-induced aging. Since the bone marrow is extremely sensitive to radiation and its failure has been shown to be responsible for some of the early radiation deaths, it might be expected to be important in radiation-induced aging. The fact that it is not eliminates the possibility that the hematopoietic system is responsible for this phenomenon.

Next, an investigation was undertaken of toxic agents other than radiation which might cause a shortening of the life span. A current theory postulates that aging is an accumulation of the effects of stress, each causing a certain amount of irreparable damage. Therefore, large single doses of several noxious agents, including typhoid toxoid, tetanus toxoid, and nitrogen mustard, were administered to young mice, which were then

allowed to live until natural death. These doses caused no shortening of the life span in these animals, although comparable doses of radiation caused a marked shortening.

The implications of this experiment were considered so important that it was decided to check the experiment in every possible way. Consequently, nearly two years ago a drastic series of treatments was started in which noxious agents were administered to mice either weekly or bi-weekly for almost their entire life span. The agents used were typhoid toxoid, typhoid toxin, tetanus toxin, tetanus toxoid, nitrogen mustard, and turpentine. These experiments are now nearly complete and the results seem clear. Provided the treatments are not so drastic as to kill acutely, the mice live as long as the untreated animals. Thus it seems evident that a nonspecific stress of the types employed so far is not life-shortening, as is radiation; hence it is necessary to look for some other mechanism by which radiation shortens the life span.

Work is continuing on the equivalence between x-rays and neutrons as regards radiation-induced aging. It has been shown that in fairly large doses, of the order of 30% of that necessary to cause death acutely, x-rays have the same efficiency as neutrons for both short-term and long-term effects. This confirms results reported from other laboratories for large single doses of these radiations. However, for chronic x-ray or neutron irradiations the ratio of efficiencies has been reported to be quite different. Thus, there seems to be a marked difference in the aging effect, depending on whether the radiation is given in a single large dose or as many small doses over a long period of time. An analysis of available data indicates that aging by neutrons depends only on total dose received and is independent of dose rate, while for x- or  $\gamma$ -rays the aging effect is very dose dependent. With x-rays a single large dose appears to be as much as eight times as effective as an equal but divided dose given over a large fraction of the life span. This concept is being subjected to further experimental test.

The work described above was carried out with the mouse as the experimental animal. Extension of such study to other animals is desirable; therefore a pilot study using the rat has been made. With doses of 500 to 800 r of x-rays or neutrons, most of the animals die either before 90 days or after 300 days. The relatively early deaths are in

part the result of effects on intestine or bone marrow, and they increase linearly with dose of radiation. In contrast, the relatively late deaths are attributable to no specific cause, they increase faster than the dose of radiation increases, and thus may be taken to represent radiation-induced aging. An effect of irradiation on life expectancy is apparent in the rat with doses of  $\approx 100$  r. With lower doses the results are equivocal; at the 25-r level a slight increase in life expectancy may occur.

#### **Acute Effects of Irradiation on Metabolism**

Earlier it was found that in the rat the effects of whole-body irradiation on carbohydrate metabolism, indicated in part by an increase in blood glucose and liver glycogen, are mediated in large part through the pituitary and the adrenal glands. The order of events appears to be stimulation of the pituitary by the irradiation, stimulation of the adrenal cortex by material from the pituitary, and then action of adrenal steroids from the cortex on carbohydrate metabolism. Removal of the pituitary or the adrenals prevents the effects of the irradiation.

It now appears that irradiation produces effects on nitrogen metabolism that are under similar hormonal control and are probably related to the effects on carbohydrate metabolism. Intact rats invariably have an increased nitrogen elimination after irradiation, whereas adrenalectomized animals show no such increase. In both intact and adrenalectomized rats, irradiation is followed by a decrease in the weight of the thymus and spleen and an increase in the weight of the liver. It appears probable, therefore, that protein from the radiosensitive tissues, e.g., the spleen and thymus, is transferred in part to the liver as amino acids. If the animal is intact or if adrenal extract is given to the adrenalectomized animal, the amino acids are converted by the liver to glycogen, and the resulting nitrogen is excreted in the urine. In the absence of the adrenal glands, the amino acids are virtually not converted to glycogen but are stored as amino acids or as protein.

#### **The Effects of Radiation on the Intestinal Epithelium**

In animals irradiation has long been known to damage the intestine grossly. The damage is manifested by nausea, vomiting, diarrhea, and limited

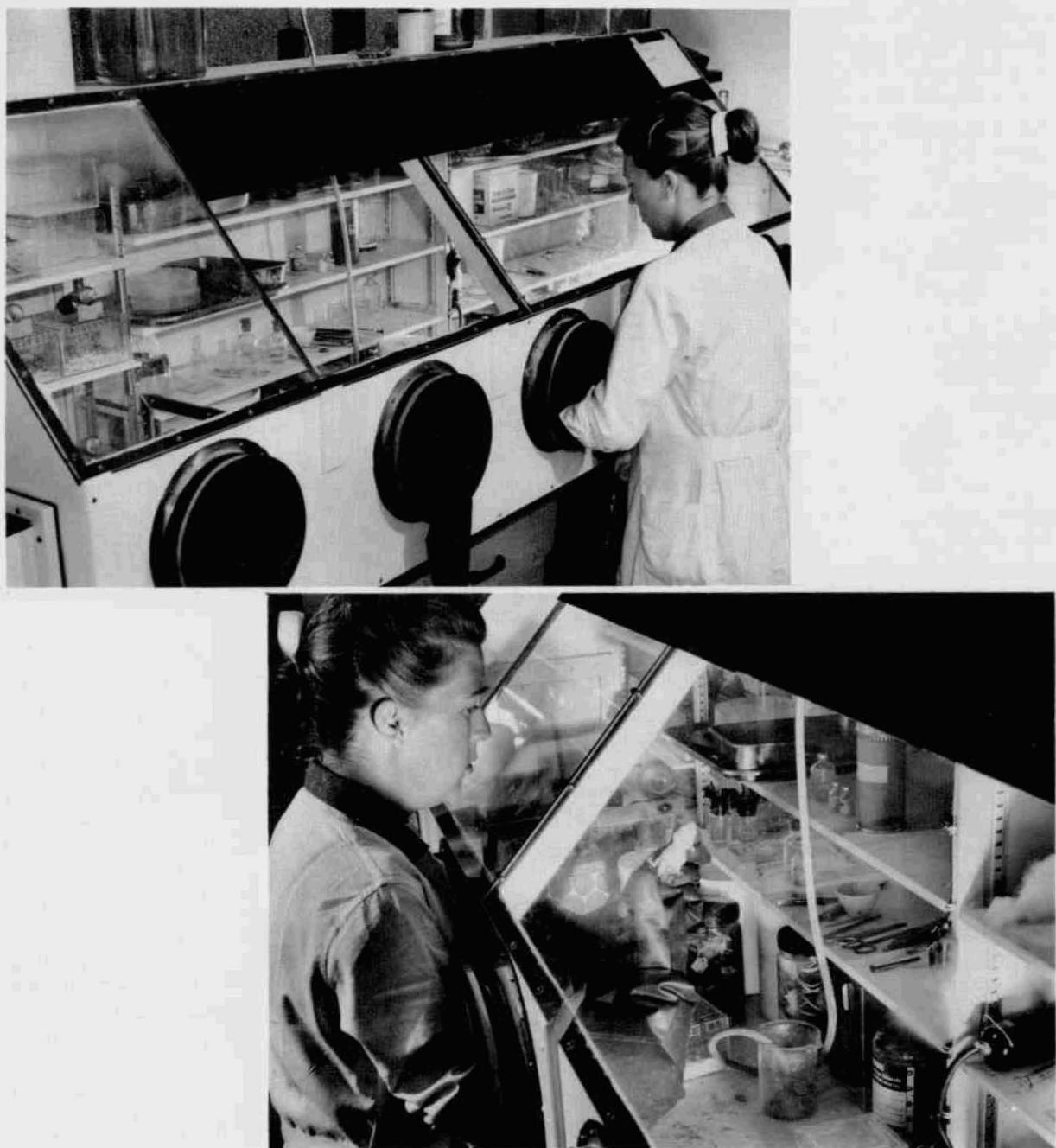


Figure 1. Small, self-contained laboratory and animal quarters for use in experiments with tritium-labeled thymidine and other radioactive materials. The labeled thymidine has proved of great value in the study of the synthesis of deoxyribose nucleic acid, a constituent of chromosomes, and in the study of chromosome formation during cell division. Although the radiation from the tritium is weak, a property that makes it uniquely suitable for localization of labeled material by autoradiography, the concentrations required are relatively high and potentially dangerous, particularly when the tritium is a component of material of long biological lifetime, such as thymidine. Experimental animals, equipment, and other materials are introduced at the left. During the course of an experiment, the operator works with gloved hands within the box. Contaminated and used material and material for further study are removed from the box through a door at the right. A slow stream of air is drawn through the box at all times.

food intake. With sufficient radiation, failure of the intestine is a prime cause of death.

It is clear that the damage to the intestine involves loss of epithelial cells. These cells are known to be subject normally to steady loss and replacement, and thus represent a cell population in a state of dynamic equilibrium. Consequently, study of the nature and causes of the loss of the epithelial cells under radiation has a bearing on the kinetics of other such populations.

Study of the effects of radiation on the epithelium has revealed that the initial effect is upon cell production. This leads to a decrease in the number of cells lining the intestine. If the generative cells recover, they produce new cells at a greater than normal rate and thus repopulate the tissue. If the generative cells do not recover, the rate of decay of the epithelial cells decreases to less than normal, and the cells form a thin layer that persists for some time. This persisting thin layer, however, is not sufficient to prevent passage of bacteria and later death. Under relatively low intensities of continuous irradiation, adaptation can occur and a new steady state is established. The cell population which produces new cells maintains its size, but fewer cells are produced and many are not normal.

Collaborative work with members of other institutions and the Medical Department has yielded further information. In this work thymidine labeled with tritium was used, and autoradiography was employed to locate the tritium. The thymidine is incorporated into deoxyribose nucleic acid, a component of chromosomes, during the synthesis of the acid that precedes cell division. The labeled nucleic acid is retained by non-dividing cells until their death. Thus the fate of single cells can be followed.

Normal and irradiated animals were given the labeled thymidine, and the location of labeled epithelial cells in the intestine was determined after suitable intervals. The kinetics of the normal cell population was established in appreciable detail for a particular strain of mouse: Nucleic acid synthesis is terminated about an hour after the onset of mitosis. The doubling time for the acid is about 7 hr, and the interval between the onset of synthesis and preceding mitosis is about 10 hr. Cell production takes place in the crypts of the intestine. During their active life, the cells migrate from the mouths of the crypts to the tips of the villi. The progress of the labeled cells indicates

that this process takes from one to two days, depending upon the section of intestine studied.

Comparison of the findings for the normal and irradiated animals gives rise to the following concept of the effects of radiation on the intestinal epithelium: The irradiation prevents resting cells from entering the phase during which the nucleic acid is doubled. Cells in the phase of nucleic acid synthesis are inhibited; presumably the rate of synthesis is decreased. Some of the cells that proceed through the nucleic acid doubling phase are unable to divide. Each of these attacks by radiation leads to a decrease in cell production. The migration of the cells formed under radiation from the generative zone to the surface does not seem affected; however, the cells show a variety of abnormalities. The analysis makes it clear that inhibition of the process leading to cell division (mitosis) by irradiation is much more profound than the inhibition of nucleic acid synthesis.

#### **Information Theory of Radiation Mortality**

A number of formulas exist which relate radiation mortality to the amount of radiation received. Some have a theoretical basis, others do not, and all are valid over restricted domains. A formula has been developed which has a theoretical basis and apparently far-reaching validity. The formula, based on information theory, is obtained by the development of a known relation between errors in interpretation of a whole message and distortion of its parts into a general relation between component perturbation and system failure. This formula relates mortality rates to two factors, one being simply a scale factor, the other a measure of organizational stability. The relation has the predicted form in the following situations: acute radiation mortality (many species); late mortality after single, repeated, or continuous irradiation; and tumor incidence. The relation also applies to other situations, e.g., general degenerative changes and, probably, effects of noxious agents.

#### **Hormonal Control of Carbohydrate Metabolism**

The hormonal control of mammalian carbohydrate metabolism has been under continued study in collaboration with groups at Columbia University and New York University. The specific technique involved is the intravenous injection of extremely small amounts (by weight) of glucose containing C<sup>14</sup> to label the blood glucose in transit from the liver to cells of other tissues.

A question of great current interest is whether or not insulin directly inhibits glucose production by the liver. Results obtained elsewhere by use of  $C^{14}$ -glucose have been interpreted to mean that glucose production by the liver is stopped by injected insulin and also by the endogenous insulin secreted by the pancreas in response to a high glucose level in blood. An extension of this interpretation leads to the conclusion that the direct action of insulin on the liver cells to reduce their output of glucose is of importance in correcting the symptoms of diabetes.

Work carried out here revealed that the apparent cessation of unlabeled glucose production by the liver observed by others during the temporary hyperglycemia induced by injection of a large amount of unlabeled glucose is an artifact brought about by the slow mixing of the injected glucose with a portion of the body glucose. When the injected load of glucose was labeled to match the level of the label in the body glucose, this mixing error was eliminated, and the continuing dilution of the circulating  $C^{14}$ -glucose by untagged glucose from the liver was apparent during hyperglycemia. Thus the insulin presumably secreted by the pancreas under the influence of the high level of blood glucose did not appreciably inhibit the output of glucose by the liver.

The interpretation of the rate of  $C^{14}O_2$  expiration by the animal metabolizing  $C^{14}$ -glucose is complicated by the "dead space" of body carbonate interposed between the  $CO_2$  arising in the tissue cells and the  $CO_2$  expired by the animal. A theoretical treatment of data developed here takes this dead space into account and thus allows interpretation of  $C^{14}O_2$  production. The application of the treatment is indicated by the following: By the end of the 5-hr test period,  $\approx 50\%$  of the glucose taken up by the tissues in both the normal and the adrenalectomized dog appears as expired  $CO_2$ . However, in the adrenalectomized animal the expired  $CO_2$  from the glucose appears "directly." The remainder of the glucose is retained as tissue constituents that do not appear as  $CO_2$  during the test period. In contrast, in the normal dog 40% of the glucose taken up by the tissues appears directly as expired  $CO_2$ , and another 10% appears during the course of the test period.

The relative importance of 11,17-oxy corticosteroids from the adrenal gland (e.g., cortisone) and the growth hormone from the pituitary in maintaining normal glucose metabolism is of con-

siderable moment. It has been found that in the undisturbed postabsorptive state the effects of a lack of growth hormone are more easily discernible. However, in the animal subjected to fasting or to insulin hypoglycemia the effects of a lack of adrenal steroids become apparent.

## GENETICS

### Radiation-Induced Somatic Mutations in Plants

One area of study pertaining to the effects of radiation on plants is the induction of somatic mutations. Production of such mutations is the basis of efforts to improve agricultural and horticultural plants by irradiation. Understanding of these mutations may lead to refinement of the methods employed in attempts to produce useful kinds.

The expression of somatic mutation currently under consideration is change in flower color. Experimentally, change in color lends itself to ready detection and analysis. Under long-term gamma irradiation the flowers that develop on the plants tested, *Antirrhinum*, *Petunia*, *Tradescantia*, and *Lilium*, bear relatively large numbers of small spots of changed color which represent somatic mutations. The mutations are also less frequently expressed as comparatively large sectors and in a few cases as whole-flower or whole-branch changes in color.

With short-term x-irradiation in *Antirrhinum* and *Petunia* many small spots are induced simultaneously. They occur as a fairly constant proportion of the cell population during the last ten cell generations in maturing petal tissue. Thus, the frequency of the spots seems a valid measure of the frequency of induction of the mutations by irradiation. With short exposures of *Antirrhinum* flower buds to x-rays, the mutation rate is increased from the spontaneous level of  $\approx 50$  per million cells by an average of 2 per million cells per roentgen increase in dose.

In all the plants studied the mutation rate increases linearly with dose of radiation. Under gamma radiation the mutation rate is highest for *Lilium*, and it is higher for *Tradescantia* than for the other two plants. The range is about 200-fold.

While many factors can influence mutation rates, the relative responses of the plants indicate that the rate of production of the mutations by irradiation is a function of chromosome size. This suggests that the mutations arise from chromosome aberrations rather than from point effects.

In contrast to the mutant material represented by the small spots and sectors on the petals, that involving the whole flower or branch can be propagated vegetatively. However, these large sectors are periclinal chimeras with mutant epidermis and nonmutant internal tissue. Hence, effective genetic test for the nature of the mutations cannot be made. It is presumed that they represent greater or lesser deletions of chromatin, including the dominant gene for flower color.

In most instances flowers that form and develop after the plants are no longer subjected to irradiation show no mutations. However, spontaneously mutable stocks of *Antirrhinum* have been obtained from irradiated material. Such material arises more frequently than the relatively rare whole-flower or branch mutations. When subjected to irradiation some of the spontaneously mutable types show an increase in mutations while others do not. The nature of these mutable stocks is not yet known.

The relation between the particular form a mutation of the kind under consideration will take and the development of the flower is not completely clear. If the mutation occurs at the earliest stage of development, expression as a large sector might be expected. However, the mutant cell may not have the capacity to establish itself and its progeny as leaders in the growing point; also, the mutation may be associated with inviability. Thus, large mutant areas might be expected to be, and indeed are found to be, rarer events than the small spots. The latter presumably arise from mutations that occur after differentiation of petal tissue at the growing point is well under way.

#### Genetic Studies on Tumor Initiation in Plants

Although a relation is indicated between tumor initiation and somatic mutation, little or no experimental testing has been done in the same plant. In order to study this relation, attempts were made to synthesize plants on which both phenomena could be observed. The amphidiploid *Nicotiana suaveolens-langsdorffii* was crossed with *N. sanderae* to form an interspecific triploid in which the combination of the first two species normally produces spontaneous tumors, and the third contributes a single dominant allele (*P*) which gives a red flower for scoring "mutation." Unexpectedly this hybrid did not form tumors; the presence of a genome from *N. sanderae* prevented tumor formation.



Figure 2. Whole-branch somatic mutation produced in *Petunia* by gamma irradiation (150 r/day during 58 days). The dark flowers bear the normal color. The white flowers bear the mutations. All the flowers on one branch are white.

To retain the tumor-forming property of *langsdorffii* combined with the pigment factor from *sanderae*, use was made of cultures derived from a cross of *N. langsdorffii* with *sanderae* which had been backcrossed four times to *langsdorffii* and selected for *P*. These were crossed with *N. suaveolens* and with *N. suaveolens-langsdorffii*. The progeny all formed tumors and had red flowers bearing the *P* allele.

Three lines so derived were grown under normal field conditions and under gamma radiation given in doses ranging from 1.24 to 200 r during a daily 20-hr period. Correlations between radiation dosage and loss of *P* were essentially linear, but the "mutation rate" for *P* differed significantly among the lines which had somewhat different residual genotypes. The increase in somatic mutation rate over that for the controls was  $\approx 100$ -fold at 200 r per 20-hr day.

In these same plants the frequency of tumor initiation was determined by noting the percentage of nodes with tumors on the primary stem, three secondary branches, and twelve tertiary branches. There was an over-all correlation between age of tissue and tumor frequency in control and irradiated cultures: on the primary stem 81% of the nodes were tumorous, on the secondary branches 76%, and on the tertiary branches 62%. No correlation was found between frequency of nodal tumors initiated and radiation dosage or frequency of somatic mutation. It appears, therefore, that there is no simple relation between tumor initiation and somatic mutations. Further studies are in progress with clonal populations and a wider variety of *Nicotiana* genotypes grown in a greater range of radiation dosages to test the general validity of the results so far obtained.

Since a genome of *N. langsdorffii* combined with *N. suaveolens* (or *N. glauca*) causes tumor formation, but *N. sanderae* prevents tumors, progeny from the first generation of the cross of *langsdorffii* and

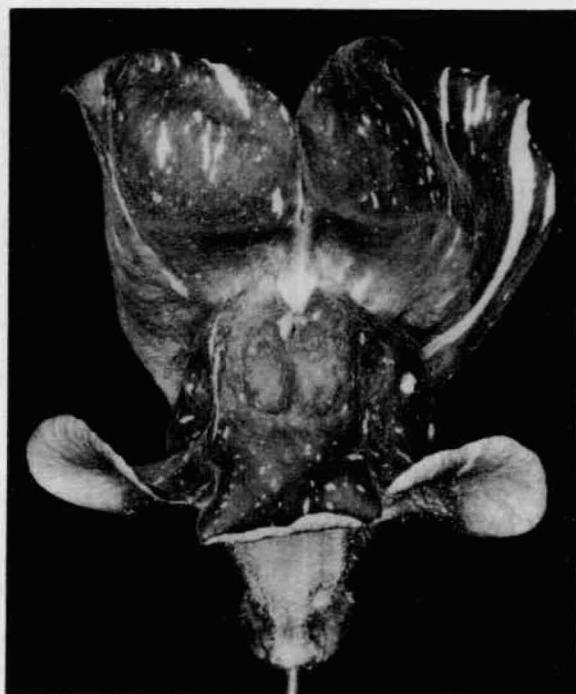


Figure 3. Somatic mutation produced by irradiation of *Antirrhinum* (Snapdragon). The light areas on the flower represent mutations. Usually flowers that form and develop after the plants are no longer under irradiation show no mutations. The flower in the photograph is from a plant that continued to exhibit mutations after irradiation.

*sanderae* when crossed with *N. suaveolens*, *N. suaveolens-langsdorffii*, *N. suaveolens-glaucum*, or *N. glauca-langsdorffii* would be expected to segregate for genetic factors affecting tumorization. A small population of the first cross confirmed this expectation. Large populations of each cross are now being grown to study further the genetic control of spontaneous plant tumors and their response to radiation.

#### Abnormal Growth and Tumor Induction in Plants

Some 5 to 6 weeks after high doses of gamma-rays given at fairly high rates, small protuberances appear on the surface of the leaves of *Graptopetalum paraguayense*. The protuberances seem to arise from new, tumorlike growth centers which are distributed at random and are apparently self-limiting in size. At maturity the tumors consist of a broad central region surrounded by several rows of cells. This complex is usually surrounded by a region of cell enlargement which extends into the normal mesophyll tissue. The maximum number of tumors obtained was about ten per leaf. Since the tumors can be counted readily and since metastases do not occur, the material offers certain advantages over others for study of tumor induction by radiation.

Any appreciable understanding of radiation-induced tumors requires information about other kinds. A preliminary test seeking such information indicates that plants derived from a cross of *Nicotiana glauca* and *N. langsdorffii*, a combination susceptible to both spontaneous and radiation-induced tumor formation, when treated with triiodobenzoic acid develop tumors of a type similar to those produced by irradiation but different from those known to develop spontaneously. The relation of these tumors to those produced by irradiation is not at present apparent; however, triiodobenzoic acid is considered to inhibit auxin transport, and irradiation appears to affect auxin metabolism.

#### The Use of Genetic Markers in Cytogenetic and Morphogenetic Studies

The survival of seedlings from irradiated seed, like the survival of other irradiated organisms, decreases with increasing dose of radiation. However, in contrast with the survival of many other organisms, that of seedlings falls off abruptly with increase in dose of radiation beyond some given point. Results obtained with corn embryos bear on

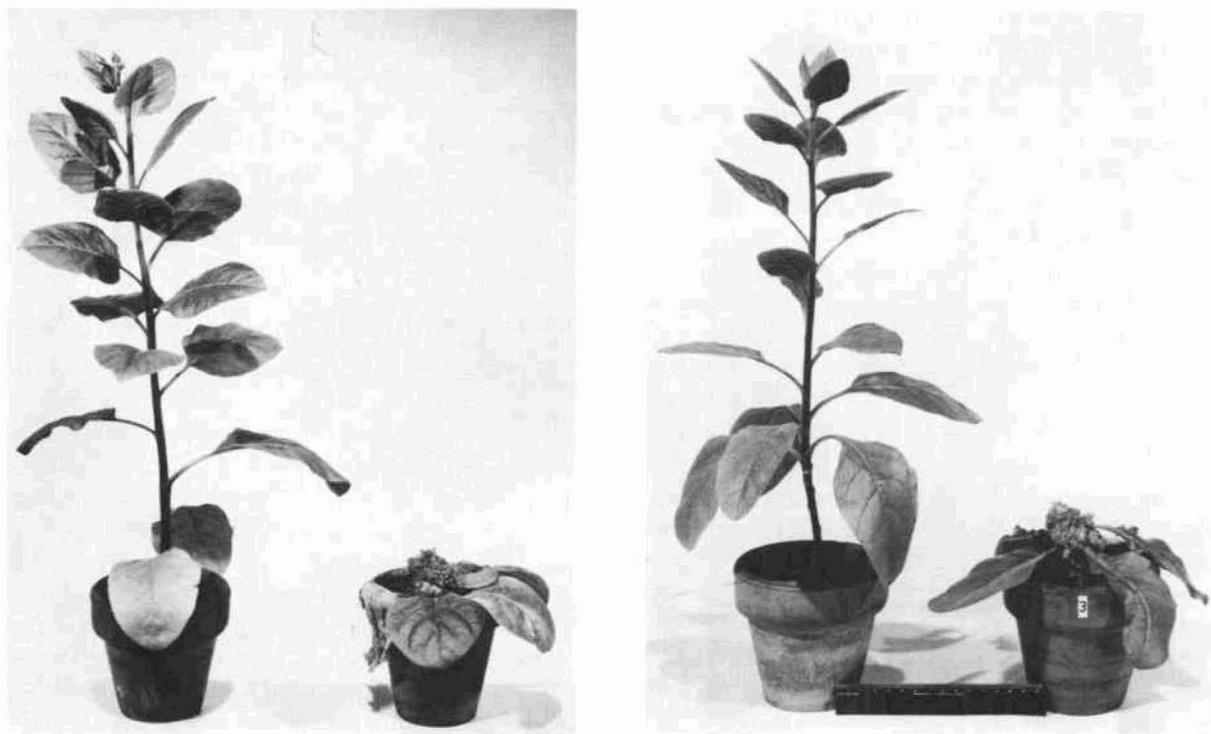


Figure 4. Tumor formation on the cross of *Nicotiana glauca* and *N. langsdorffii*. The second plant from the left bears a tumor that developed 6 wk after treatment with 2700 r of x-rays. The plant at the extreme right bears a tumor that developed 4 mo after treatment with triiodobenzoic acid. This tumor is like that produced by the irradiation. On the left in each case is an untreated control plant.

the abrupt decrease in survival and illustrate a procedure for assessing the maximum genetic damage compatible with survival. Corn embryos heterozygous for the gene yellow-green ( $yg^2$ ) were soaked in water for 24 hr, irradiated, and then allowed to develop. The length of roots and shoots during seedling growth and the number of yellow-green sectors on the leaves were measured. The yellow-green sectors derive from mutation; presumably they result from loss of the dominant gene on the end of chromosome 9.

There was fair correlation between the loss of genetic material and reduction of growth. Although without direct bearing on the problem under discussion, it is of interest that low doses of indole acetic acid under certain conditions will increase the height of the experimental seedlings to that of controls. The leaves that were morphologically distinct at the time of irradiation (leaves 1 to 6 in the stock used) showed a linear relation between number of yellow-green sectors and dose of radiation. Leaves 7 to 9, which are identifiable as

particular portions of the growing point, responded in essentially the same manner as leaves 1 to 6. Leaf 10 and those beyond, which represent new growth resulting from cell division within the apex, responded linearly through a limited range of dose and then showed no further increase in number of sectors with increase in dose.

It appears from these results that the cells at the apex can carry only a limited load of genetic damage. With further damage to the apex, leaves do not appear, and the seedling does not survive. The results also imply that the effect of irradiation on plant growth is not explainable by attack on single cells, but involves some minimal mass of an "organizer" region of the apex. This concept explains the relatively all-or-none response in seedling growth and survival.

#### Self-Incompatibility in Plants

Self-incompatibility in plants signifies the inhibition of pollen germination or of pollen tube development and, consequently, seed formation

that occurs when certain plants are self-pollinated. The process is under genetic control of the *S* locus. Self-incompatibility (*S*) studies have been focused on three problems, namely, those of *S* locus structure, *S* gene action, and *S* gene distribution in the plant kingdom.

For the elucidation of *S* gene structure, x-irradiations have been applied to flowers and buds of five homozygotes and three heterozygotes of *Petunia inflata*. Pollen from 3500 treated (250 to 1000 r) and control flowers was applied to flowers of untreated clones of the same genotype. Studies of the progeny from these matings, which included both self-fertile and self-incompatible plants, were then made. Considering all data, irradiation treatments resulted in a threefold increase in "mutant" seed production. More striking, however, was the nine-fold increase in seed from heterozygous vs that from homozygous clones (similar in control and treated material). It is suggested that this increase due to heterozygosity alone is the result of radiation-induced crossing over or interchange within a compound *S* locus.

*S* gene action in plants with a gametophytic incompatibility system is interpreted to include three basic steps, namely, the production of *S* gene products in pollen and in pistil and the interaction of these two products during incompatible pollen tube growth in the style. The absence of a detectable increase in "mutants" from x-ray treated postmeiotic buds would seem to indicate that *S* gene action in the pollen occurs shortly after meiosis. The time of stylar gene action can be studied in but a few plants, i.e., those having receptive immature stigmas. Studies of three such plants (broccoli, *Nicotiana*, and *Petunia*) indicate that *S* gene action occurs rather abruptly at a late stage of flower development, and in at least one of these species it appears possible to disrupt this action by propitiously timed stylar irradiation. The final step, the interaction of *S* gene products, has been studied by means of pollen tube growth *in vitro*. Inhibitions specific to known alleles appear to be demonstrable *in vitro* with *Petunia*, and attempts are being made to isolate and purify these specific inhibitors.

Two major *S* gene systems, the gametophytic and sporophytic, occur among flowering plants and differ primarily in the time of *S* gene action in the pollen. It has been observed that gametophytic species generally represent plant families having binucleate pollen grains while sporophytic species

have trinucleate grains. The incompatibility is expressed as pollen tube inhibition in the former and as the inhibition of pollen germination in the latter, which suggests that final *S* gene action in the pollen follows the second mitotic division. These correlations have been extended with particular reference to differences in physiology and in the phylogenetic distribution of the binucleate (primitive) and trinucleate (derived) pollen states.

#### Induced Mutations for Pathogenicity in Fungi

Although it is well known that mutation effects changes in the relation between plants and pathogenic fungi, theoretical and experimental detail upon which to base practical applications is far from ample.

Induced mutations for pathogenicity have been reported for a number of plant-pathogenic fungi, but in most instances the genotype of the wild-type character was unknown, and the screening tests included only a limited number of cultures characterized primarily as biochemical or morphological variants. As a consequence, quantitative information concerning the mutation rates of single genes for pathogenicity is lacking. The work described herein concerns the dose-frequency kinetics of mutation at the *A<sub>M</sub>a<sub>M</sub>* locus in race 1 of *Melampsora lini*, a fungus causing the flax rust disease. Avirulence on the flax variety Dakota is dominant to virulence. Alteration or deletion of the dominant allele (*A<sub>M</sub>*) by irradiation of urediospores conditions virulence on the normally immune Dakota.

Information concerning a chromosome mechanism of mutation was derived from analogy of the dose-frequency kinetics of chromosome aberrations in higher plants. The mutation frequency increased directly (single-event process) with dose in ultraviolet light (UV) treatments, but increased as the square (mutation resulting from a two-event process) of the dose in x-ray and  $\gamma$ -ray treatments. Near linearity in the neutron dose-frequency curves, contrasted with curvilinearity for x- and  $\gamma$ -rays, is attributed to differences in ionization density. The relative biological effectiveness (rbe) of neutrons versus x-rays ranged from *ca* 22 at the lowest dose to 14 at a high dose level. The ratio of the maximum frequency induced by fast neutrons, x-rays, and UV was *ca* 7:5:1. Approximately the same ratio was obtained for several other loci in a recent experiment. Loss of growth vigor, an indication of associated genetic damage, was less ap-

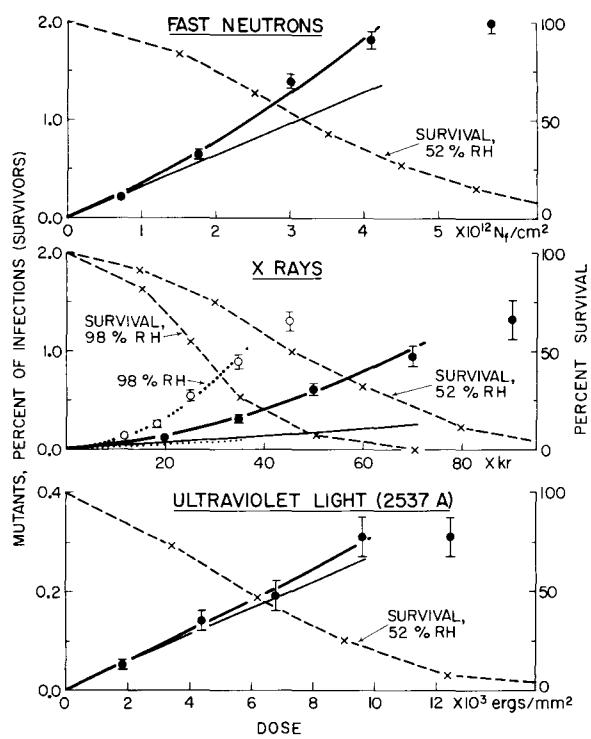


Figure 5. The relation between the dose of radiation and frequency of mutation (based on infections of flax, Dakota variety), at the  $A_{Ma_M}$  locus for pathogenicity in the fungus *Melampsora lini*. The experimental points are fitted to the polynomial,  $F = bx + ex^2$ , by the method of least squares. The straight lines represent the linear component of the heavier curvilinear lines. RH signifies the relative humidity during the irradiation. The frequency of mutations at 50% survival is somewhat greater with neutrons than with x-rays, and each of these radiations is much more effective than ultraviolet light.

parent in UV-induced mutants than in those induced by the other (ionizing) radiations. From these considerations it is suggested that mutations to greater virulence in *M. lini*, induced by ionizing radiations, result largely from chromosome deletion rather than gene mutation. More localized ("point") changes, or simple deletions resulting from single chromosome breaks, are indicated for UV treatment. In this instance, loss of a character (avirulence) increased rather than decreased the survival probability of the organism. The relative importance of the deletion vs point effects in spontaneous mutation in nature is as yet unknown. The role of such "spontaneous" mutation, relative to other genetic processes which may contribute to the origin of new, virulent biotypes, is also unknown. The high induced rates for single loci and the prevalence of potentially mutagenic agents

(especially solar UV and toxic chemicals in necrotic plant tissue) suggest a major role for "spontaneous" mutation. From a practical standpoint, the relative induced mutability of the various genes for pathogenicity in this fungus will aid the plant breeder in evaluating the usefulness of the corresponding genes for resistance in the host varieties.

A number of chemicals have been employed in preliminary tests for mutagenicity at the  $A_{Ma_M}$  locus. The dose-frequency response to nitrogen mustard, the most effective chemical mutagen used, is similar to that of the UV treatments. As in the radiation treatments, a "saturation" level (peak, followed by a decline) in mutation frequency occurred at the high doses.

#### Lysogeny and the Genetics of Bacteriophage

Infection of bacteria by temperate bacterial viruses or bacteriophages may produce either of two results. The infected cell may be destroyed after a short latent period, liberating hundreds of progeny phages; or the host may survive the infection, giving rise to a hereditarily changed bacterial clone. The surviving clone is immune to subsequent infection by the phage and can produce the phage in the absence of fresh infection. The infecting phage disappears as such, but its genetic material is integrated into the bacterial genome and provides the genetic control for the hereditary changes. The altered state of the bacterium is known as lysogeny, and the term prophage is used to designate the controlling entity of the lysogenic condition.

The wild type of phage P22 of *Salmonella typhimurium* is temperate. A large number of spontaneously arising mutants of P22 affecting the ability to lysogenize have been examined. Phenotypically all such mutants fall into three groups. One group of mutants is still temperate, but lysogenizes somewhat more poorly than the wild type. The mutants of the other two groups fail to lysogenize by themselves. However, if bacteria are infected with mixed phage of both these groups, lysogenization occurs. That is, two mutants, each unable to lysogenize, are able, when put together in the same cell, to interact physiologically to give the lysogenic condition, with one of these mutants as prophage. This phenomenon has been called cooperation.

Genetically all the mutations fall into a circumscribed region of the phage linkage map. The

three classes of phage mutations affecting the ability of the phage to lysogenize fall into three distinct and closely linked pseudoallellic series or functional units. The mutations of any one class can be mapped in a single unit.

It has been assumed, as a working hypothesis, that a number of reactions are necessary for lysogeny. Each group of mutants is blocked in the ability to carry out one of these reactions. Two mutants belonging to different groups, presumably blocked at different steps, when put together in the same cell can cooperate to complete all the reactions necessary for establishment of prophage.

A number of environmental treatments affect the ability to lysogenize. Ultraviolet (UV) irradiation of phage before infection decreases the probability of lysogeny. This effect of UV is photoreactivatable to some extent. Similarly, treatment of infected bacteria with the antibiotic chloromycetin reverses the UV effect, even when given very late in the latent period. Chloromycetin is said to be

an inhibitor of general protein synthesis. These findings suggest that a late-forming protein may be involved in the decision between lysis and lysogenization of bacteria infected with temperate phage. A study of the interplay of these treatments with each other and with the phage mutants affecting the ability to lysogenize may shed light on the reaction steps postulated above.

A striking effect of low temperature on the UV sensitivity of phage P22 has been discovered. Phage P22 is 5 to 10 times as sensitive to a given dose of UV irradiation when frozen in dry ice and acetone as at room temperature. Both treatments are susceptible to photoreactivation to the same extent. The possibility of more efficient migration of energy in the frozen state, possibly between the DNA and the protein of the phage, is being investigated.

Experiments utilizing radiosotopes and the techniques of autoradiography have recently been started on the ciliated protozoan, *Paramecium aurelia*. Under investigation now is the "killer" character. Killer paramecia contain a desoxyribose nucleic acid plasmagene which produces a poison, paramecin. This poison is excreted into the medium and there picked up by sensitive animals. One particle of this poison can kill a sensitive cell. It is hoped to label paramecin and follow its mode of adsorption to the site of action in sensitive animals.

#### Cooperative Radiation Mutation Program

The radiation facilities at Brookhaven – in particular the field in which plants can be grown under long-term gamma radiation, the neutron source at the reactor, and the x-ray machine in the Biology Department – are being used to irradiate plants, seeds, cuttings, and the like for eventual study elsewhere. The goal of this work is to assess the usefulness of such irradiations in producing mutational changes that represent crop improvements. Irradiations have been performed for many individuals and groups in this and other countries.

Reports within the past year from several co-operators disclosed the production of new mutations of agricultural significance. Scientists at Rutgers University, who previously have reported mutations for early and late ripening in Fairhaven peaches, have found a late-ripening branch in the important peach variety Elberta. Fruit on this branch consistently ripens two weeks later than that on the rest of the tree. Associated with this

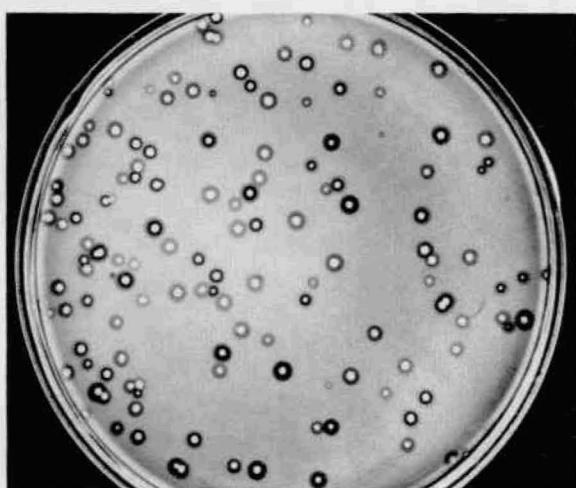


Figure 6. Agar plate showing four mutants of bacteriophage P22 of *Salmonella typhimurium*. The four types were obtained by allowing a mixture of two to infect the bacteria and multiply. The phage progeny were recovered, mixed with bacteria, and placed on the plate. Each plaque on the plate represents the action of a phage particle. The clear center of each plaque is an area in which the bacteria were lysed by action of the phage. The circles and halos indicate changes in the bacterial colonies characteristic of the phage mutant. One parent type of progeny produced the plaques with a dark ring; the other parent type produced the plaques with a light ring surrounded by a marked halo. One derived type of progeny is represented by the dark ring and a marked halo, the other by the light ring. The relative number of each type is that expected for a cross of the kind made.

late-ripening character is greater firmness of the fruit flesh, which increases the value of the fruit for shipping purposes. In the peach variety Brackett a mutation was found from the usual freestone condition to the clingstone condition. This mutation confers certain advantages on the fruit that make it desirable for special commercial purposes.

Other work by the Rutgers group has revealed a marked qualitative difference in the response of peach embryos, cultured *in vitro*, to thermal neutrons and x-rays. Embryos produced by certain controlled crosses of peaches are often of very low viability. Fruit breeders have found that these embryos can survive if excised from fruit at an early stage and grown under aseptic conditions in tissue culture. However, many of these embryos produce stunted dwarf plants whose growth is abnormal. Radiation was used in an attempt to break this block to normal growth. The use of x-rays proved ineffective over a wide range of doses tested, but with neutrons this stunting was eliminated and normal, rapidly growing seedlings resulted. Embryos treated in the thermal column with  $2.2 \times 10^{10}$  thermal neutrons grew from 2 to 3 times as rapidly as untreated embryos. The stimulation of growth is particularly evident during the early growth stages and appears to be due to a re-

lease of inhibitory factors. An investigation of this phenomenon on a very large scale is planned for the coming year, and it is expected that this new technique will greatly increase the efficiency and productivity of the normal peach breeding research program.

At the University of Connecticut, as a result of gamma field irradiation, three new types of carnation have been produced and are ready for release to growers for yield testing under standard commercial conditions. One of these is a white recessive mutation produced by irradiating the red carnation, William Sim. This mutant appears to be considerably more vigorous in growth and flowering than the original plant and also is completely free of the red flecking which characterizes many of the existing White Sim types. A second mutation, which appeared in irradiated plants of Pink Sim, is a pink type also believed to be completely blotchless, in that it no longer forms red streaks and spots on the petals. The third mutant is a red-flowering type recovered from irradiated White Sim plants. It bears, in addition to the change in color, a different and desirable type of petal formation.

The foreign cooperative studies, begun for the most part after the first International Conference on the Peaceful Uses of Atomic Energy at Geneva,

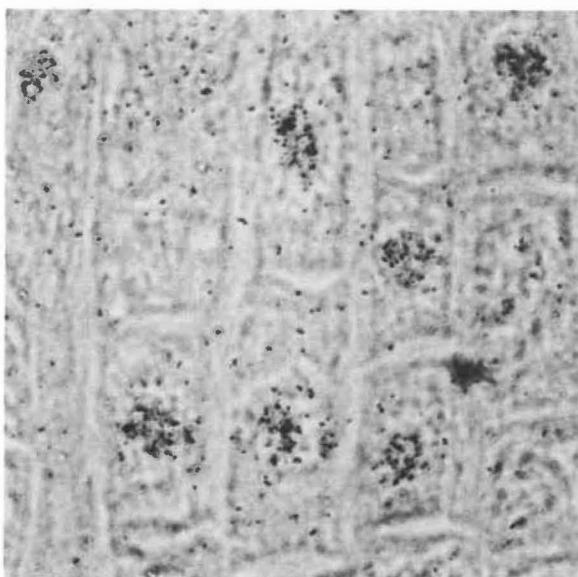
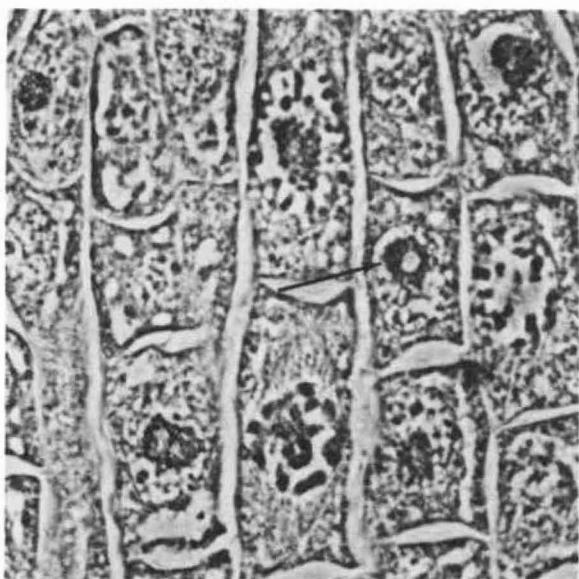


Figure 7. Accumulation of tritium-labeled cytidine in the nucleolus of the growing tip of the broad bean root. On the left the cells appear as seen in the phase contrast microscope. Note the nucleolus (arrow) contained within the nucleus. In the autoradiograph at the right, note the concentration of silver grains at the position of the nucleolus.

have continued to be the main area of the program's growth. Some of the earliest of these studies are apparently beginning to yield information, since three foreign cooperators, two from Taiwan and one from Rumania, have submitted manuscripts for the second Geneva Conference.

## CELL PHYSIOLOGY

### Nucleic Acid Metabolism in Dividing Cells

In preparation for division, a cell synthesizes duplicates of certain components, and at division each new cell contains one of the duplicates. Components of particular interest are desoxyribose nucleic acid (DNA), a major constituent of the chromosomes, and ribose nucleic acid (RNA). The DNA of the chromosomes appears to be primarily involved in heredity, the process by which specific patterns of growth, metabolism, and function are passed to progeny. RNA appears to be implicated in protein synthesis, differentiation, and embryonic development.

One way to study the metabolism of nucleic acids is to grow cells in the presence of precursors of the acids bearing radioactive elements, and then determine the location of the label by autoradiography. This involves exposing photographic emulsion to the action of the radioisotope in the cell. Microscopic examination of the cell and the apposed developed emulsion then serves to locate the part of the cell bearing the activity. Of the several radioisotopes used, tritium, an isotope of hydrogen, has given the most explicit results. Tritium is especially suitable because the effect of its radiation on the photographic emulsion is confined to the dimensions of the structures under consideration.

By using tritium-labeled thymidine, a precursor of DNA, it has been found that a chromosome consists of two entities. During cell division each entity appears in duplicate and each daughter cell contains an original and a newly synthesized entity.

In similar experiments carried out with the root-tip cells of the English broad bean *Vicia faba*, tritium-labeled cytidine, a precursor of both DNA and RNA, was used to gain information about RNA in general and the nucleolus in particular. The nucleolus is associated with the nucleus, but, unlike the chromosomes, contains RNA. The labeled precursor appeared first in the nucleolus and later in the cytoplasm. Incorporation of the label in the nuclei was variable; when present it

was associated with DNA. In other parts of the cell, the label was associated with RNA.

When cells with only the nucleoli labeled were subsequently grown in the presence of excess unlabeled cytidine, the amount of label in the nucleoli decreased, while that in the cytoplasm increased. It is possible that the newly formed RNA appearing in the nucleolus was synthesized elsewhere and transferred to the nucleolus so rapidly that the site of production could not be detected. The alternative, which seems more likely, is that formation of RNA occurred within the nucleolus. It seems clear that after accumulation or synthesis the nucleic acid was transferred from the nucleolus to the cytoplasm.

### The Structure of the Salivary Gland Chromosomes of *Drosophila*

The salivary gland chromosomes of the full-grown *Drosophila* larva are generally regarded as an aggregation of many closely appressed strands or chromonemata. This pattern of organization is presumably determined by the uncoiling, growth, and replication of the original four strands derived from the mitotic-type progenitor chromosomes found in the salivary gland of the very young larva. Presumably, this basic number of 4 increases to an eventual total of 1024 or 2048 during larval life as a result of 8 or 9 successive replications without chromosome division.

Despite the wealth of evidence favoring this multistrand or polytene concept, other interpretations of the structure continue to be advanced. All depend to a large extent on evidence gathered from observation of preparations of full-grown or developing larvae. Direct experimental evidence about the fate of individual strands in the course of chromosome growth would give a definitive picture of chromosome growth. Such evidence has been obtained by use of tritium-labeled thymidine to label the nucleic acid of the chromosomes and autoradiography to locate the labeled nucleic acid.

Larvae from hatching eggs were fed on a medium containing dead yeast and the labeled thymidine. At intervals larvae were removed and allowed to continue growth on unlabeled medium. When the larvae were mature, the salivary glands were removed, appropriately prepared, and covered with photographic film. After exposure of the film to the action of the tritium, the developed film and apposed glandular material were examined microscopically. In the larvae fed the labeled thy-

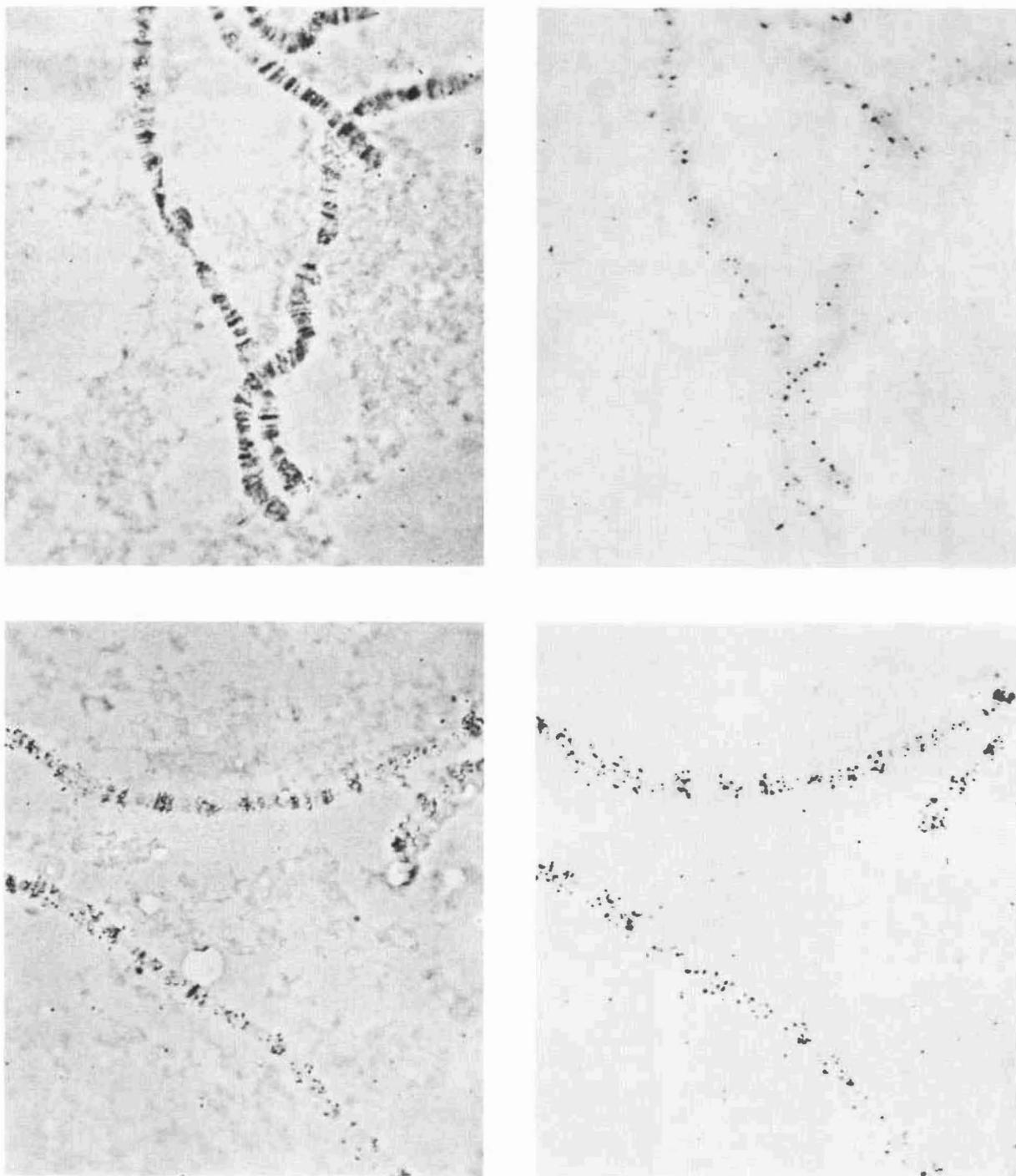


Figure 8. Structure of salivary gland chromosomes of *Drosophila* larvae. At the upper left the chromosome appears as seen in the phase contrast microscope. At the upper right is the corresponding autoradiograph. In this test a very young larva was allowed to feed on tritium-labeled thymidine for 24 hr and then on unlabeled food until fully grown. In the autoradiograph note that the silver grains appear as single strands. The lower pictures are from a test in which the larva was fed the labeled material for 48 hr. Note in the autoradiograph that more strands of silver grains occur, and that concentrations of grains coincide with the dense bands of material seen under phase contrast.

midine for a short time (24 hr), the radioactivity occurred in rows parallel to the long axis of the chromosome and traversed many bands. The longer the labeled material was fed, the more activity appeared in the chromosome. Finally, the radioactivity "filled" the chromosome. More radioactivity was found at the position of the heavier bands that parallel the short axis of the chromosome than at the position of the less dense bands or interbands. These bands, recognizable in stained and unstained preparations, have been considered to represent concentrations of nucleic acid.

The findings indicate that, as the chromosome develops, 1) new DNA occurs on a strand parallel to the long axis of the chromosome, 2) the strands are physical entities that remain intact during succeeding replications, 3) strand replication continues through a number of endomitotic cycles, and 4) the nucleic acid is not distributed uniformly along the chromosomes but is concentrated in certain regions. Thus, the multistrand concept of the chromosome is supported by relatively direct test, as are other impressions of chromosome structure.

#### The Bacterial Chromatophore and Photosynthesis

Photosynthesis, the process whereby the energy of light is used to convert carbon dioxide to sugar, involves many reactions. Broadly and in part hypothetically, these may be separated into the initial capture of the energy from light by chlorophyll; conversion of this energy into chemical energy in the form of some reductant; transfer of part of the energy to the terminal phosphate of adenosine triphosphate (ATP) through oxidation of part of the reductant; the fixation of  $\text{CO}_2$ ; and, finally, through energy derived from the ATP and reductant, the formation of sugars.

In higher plants most of the processes occur within one structural component of the cells, the chloroplast. Photosynthetic bacteria, however, contain a somewhat analogous but simpler structure, the chromatophore. This structure contains the pigments necessary for capture of light and conversion of the energy from the light to chemical energy. The other events involved in photosynthesis take place elsewhere within the bacteria. The separation of the early photosynthetic events from others in the bacteria is of considerable advantage in study of the over-all process.

The composition, structure, and function of the chromatophore has been examined in detail.

Physical chemical measurement and observations with the electron microscope indicate a spherical structure with a diameter of  $\approx 300 \text{ \AA}$ . The sedimentation constant is 143 Svedberg units. The density is 1.2485 and the "molecular" weight is about 13 million. The chromatophore has an electron-dense cortical layer  $\approx 60 \text{ \AA}$  thick and a less dense medullary region. Each chromatophore is composed of about 300 carotenoid molecules, 600 chlorophyll molecules, 300 phospholipid molecules, and protein equivalent to 40,000 amino acids. Geometric and other considerations suggest that the protein makes up the outer electron-dense layer of material, and the other constituents an inner layer adjacent to the protein.

The chromatophores convert light energy to chemical energy as indicated by synthesis of ATP from adenosine diphosphate (ADP) and inorganic phosphate in the light. Despite the apparent simplicity of composition, each chromatophore can produce about 50,000 phosphate bonds per minute. The conversion of light energy appears

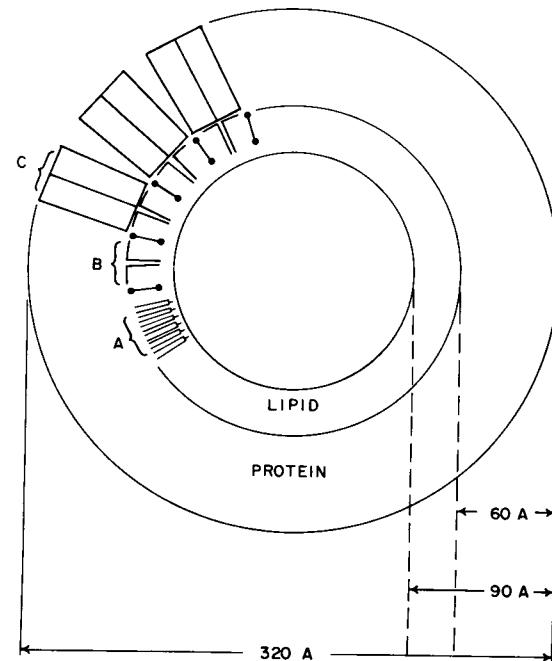


Figure 9. Hypothetical model of the bacterial chromatophore. The chromatophore is considered a sphere. *A* represents phospholipid molecules with the fatty acid chains oriented toward the periphery. *B* represents chlorophyll molecules with the porphyrin ring adjacent to the protein layer. A molecule of carotenoid pigment is between each pair of chlorophyll molecules. Geometric and other considerations suggest the arrangement indicated.

to be a function solely of the chlorophyll. The carotenoid pigments apparently serve only to protect the bacterial cell against the toxic effects of photooxidation.

### Effects of Calcium Deficiency on Plant Chromosomes

In the plants tested a calcium deficiency makes the chromosomes more susceptible to breakage by treatment with x-rays. This increased sensitivity to radiation and the more direct demonstration of the appearance of labeled calcium in nuclei indicate a close association between calcium and the chromosomes. It is not yet clear whether the calcium serves a structural or functional purpose.

The ratio of chromosome aberrations produced by irradiation in oxygen to those produced in nitrogen is the same for normal and calcium-deficient cells. Thus, the effect of calcium deficiency does not depend upon oxygen, as do the effects of many other modifiers of sensitivity to x-rays. Low concentrations of potassium are known to increase the absorption and translocation of calcium. As this effect predicts, a potassium deficiency did not increase the breakage of chromosomes by x-rays, but may have provided a small protective effect. Temperature has a profound effect on the production of spontaneous chromosome aberrations at meiosis in calcium-deficient *Tradescantia*. A 10° increase in temperature increases breakage as much as fourfold.

Collectively, these observations indicated and others suggest strongly that chromosome breakage, both spontaneous and radiation-induced, is attributable to an inherent lability of the chromosomes.

### Auxins and Root Growth

Cuttings made from the stem of the Lombardy poplar in active growth develop roots only at the basal end. In contrast, cuttings from dormant trees develop roots along the entire length. Application of indole acetic acid, an auxin, to the cuttings from actively growing trees removes the root polarization. It may be that the actively growing stems contain less available auxin than those of dormant trees. However, another explanation based on the transport of the auxin is possible. The compound is actively transported from the apical to the basal end of the cuttings, but not in the reverse direction. This unidirectional movement results in accumulation of indole

acetic acid at the basal end, and in active trees it is this basal accumulation which raises the auxin level to a point high enough to permit the roots to grow in that region.

It is possible that dormant and growing trees have equal amounts of auxin, but the rates of transport may be different under the different physiological states. If auxin is rapidly transported in actively growing stems, a deficit may exist in the apical end, and consequently roots develop only at the basal end. If transport is slow in dormant cuttings, all portions may contain sufficient auxin to allow development of roots. To test these hypotheses, cuttings from growing trees were treated with triiodobenzoic acid (TIBA), which is considered to participate in auxin activity by inactivation of the transport system. If active and dor-

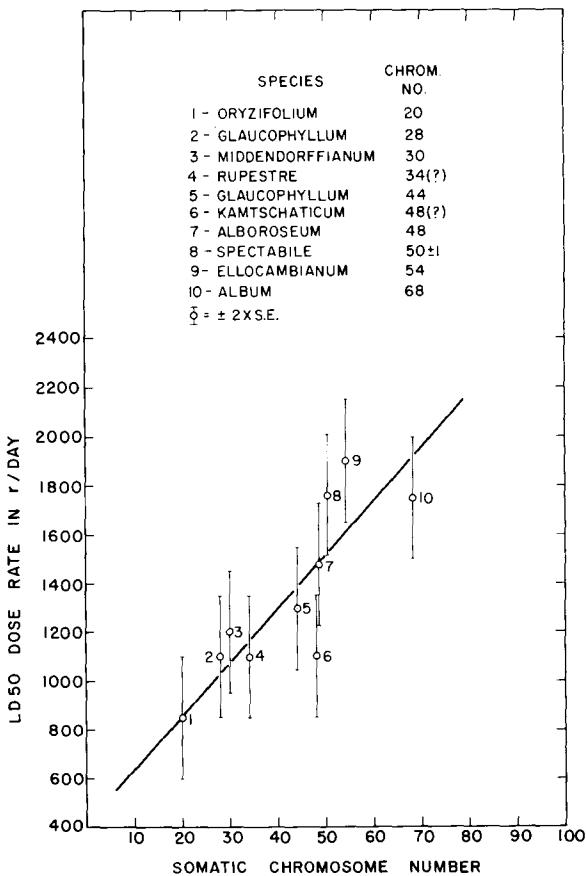


Figure 10. Relationship between chromosome number and LD<sub>50</sub> for 9 species of the genus *Sedum* after 16 wk of chronic gamma irradiation. In general, the sensitivity of plants to the lethal effects of irradiation decreases with increase in chromosome number. However, other factors, e.g., chromosome size, also influence the sensitivity.

mant cuttings have the same amount of auxin but different transport rates, the treatment of active cuttings with TIBA would be expected to produce nonpolarized root distribution. If the auxin content of active cuttings is lower than that of dormant cuttings, the treatment should prevent basal accumulation of auxin, and thus cuttings should be rootless. The second possibility obtained experimentally; treatment of active cuttings with TIBA produced viable but rootless cuttings.

#### Membrane Permeability

The major difficulty in the interpretation of coefficients obtained for transfer of water through artificial and living membrane appears to be in the concepts employed. The coefficients derive from measurements of the transfer of labeled and unlabeled water. When it is recognized that osmotic flow involves a net transfer of solvent across a membrane and that transfer of labeled water occurs by exchange with normal water, the apparent discrepancies between coefficients for osmotic flow and exchange are explained. The concept indicated allows experimental test to decide in a given case whether transfer is by diffusion or bulk flow through pores. If diffusion obtains, tracer water exchange will be little affected by a pressure gradient within the membrane. If bulk flow obtains, the exchange of tracer should depend on the gradient. The few biological membranes for which sufficient experimental data are available appear to behave as diffusion membranes.

#### The Role of Polyploidy in Determining Radiosensitivity in Plants

Since attack by radiation on cells is in part effected through action on the chromosomes, an inverse relation between radiosensitivity and number of chromosomes might be expected. The best expression of this relation might be expected to be found in related species bearing different multiples of some basic number of chromosomes (polyploidy). This expectation has been realized with a polyploid series in *Chrysanthemum* and, more recently, with the genus *Sedum*.

#### Chromosome Breakage Induced by Tritium-Labeled Thymidine

As indicated elsewhere in this report, the use of tritium-labeled thymidine to label DNA, and thus chromosomes, has provided understanding not ob-

tainable by other means. However, the radiotoxicity of the labeled thymidine has not been studied extensively. Early indications are that up to 40% of the anaphase cells in the root tips of *Tradescantia paludosa* grown for 48 hr in a medium containing 1  $\mu$ C tritiated thymidine per ml show chromosome abnormalities. The concentration indicated is not unusual in tests with labeled thymidine.

### BIOCHEMISTRY

#### Structure of Proteins

The problem raised by the relation between structure and specific biological activity reaches its ultimate complexity in the proteins. A rational exploration of the chemical architecture of the proteins was beyond experimental reach until relatively recently, when advances in methodology permitted attack on some of the very simplest and (molecularly speaking) smallest structures.

The purpose of investigations carried out elsewhere during the past five years and being continued here has been the complete elucidation of the structural formula of the enzyme, bovine pancreatic ribonuclease. This work is nearing completion. The ribonuclease molecule can be pictured as being built up by linear condensation, in an ordered and characteristic sequence, of 124 amino acid residues. The polypeptide chain formed is elaborately folded on itself by several different kinds of cross-links between certain of the residues in the chain. The spatial arrangement (tertiary structure) of the convoluted chain and of the individual residues constituting it determine the ability of the enzyme molecule to bind its substrate in the specific orientation required to bring about chemical interaction at the "active" center of the enzyme. Work in this laboratory and elsewhere is being directed to attempts to describe the action of the nuclease quantitatively in terms of the known structure, and thereby to obtain for the first time a complete description of the mechanism of an enzymatically catalyzed reaction.

It is of interest to see how far the procedures applied to ribonuclease can be applied to other proteins. Since many of the subtler manifestations of biological action reside in the tertiary structure, at present accessible only by indirect chemical means, it is hoped to couple the chemical attack with a procedure theoretically capable of reaching into this area directly. The procedure, x-ray diffraction analysis, has been used with some success

with myoglobin, a protein from muscle with the oxygen-carrying properties of blood hemoglobin. In particular, the location of the heme group has been ascertained. A combined x-ray (Dr. J.C. Kendrew and colleagues, Cavendish Laboratory, Cambridge University) and chemical attack is under way.

### The Active Site of Enzyme Action

As pointed out above, the relation between structure and biological activity is especially complex in the case of proteins, particularly those that are enzymes. The problem here, in part, is to locate among the hundred or more amino acids making up the enzyme those that are immediately involved in conferring specificity for a particular substrate and those involved in catalyzing the breakdown of the substrate. Marked insight into the problem has been obtained by study of phosphoglucomutase, the enzyme that effects the transfer of a phosphate group from one end of the glucose molecule to the other. An intermediate in the reaction is the enzyme bearing covalently linked phosphate.

The enzyme was labeled with radioactive phosphorus in the form of the phosphate group. When the enzyme was broken down by various means the phosphate stayed attached to the same amino acid (serine). By separating peptides (partial breakdown products of the enzyme) of different sizes, it was possible to determine the amino acids on either side of the one bearing the phosphate. In this way, the amino acids at the critical active site responsible for enzyme activity were determined. Of further moment, a common sequence of from three to six amino acids was found to be present in several enzymes of widely different activities. By means of irradiation experiments, histidine was also found to be present at the active site of all these enzymes. The histidine is probably located in a different but neighboring portion of the chain of amino acids making up the enzyme. This common grouping of amino acids in enzymes of very dissimilar specificities suggests that they are involved in bond breaking, and that other groupings determine the specificities.

A logical consequence of this conclusion is that the specificity of an enzyme can be changed without its catalytic activity being destroyed. Preliminary indications of this have been obtained. For example, it has been possible to develop proteolytic activity in crystalline phosphoglucomutase.

### Muscular Contraction

Muscular contraction is known to involve the contractile protein, actomyosin, and transfer of energy from adenosine triphosphate (ATP), but the manner in which the ATP transfers its energy to the protein has eluded detection. By use of water labeled with the oxygen isotope O<sup>18</sup>, a major clue to the mechanism of this conversion of chemical to mechanical work has been provided.

When a contracting preparation of actomyosin was treated with ATP in oxygen-labeled water, the inorganic phosphate derived from the ATP contained labeled oxygen far in excess of that predicted for simple hydrolysis of the ATP. Tests showed that the excess was not introduced by exchange of oxygen between the water and the initial reactant, ATP, or the final product, inorganic phosphate. Thus, the exchange must have taken place on the protein, i.e., the exchange was between the water and an intermediate formed during hydrolysis of the ATP.

From similar experiments strong indications were obtained that the intermediate was on the myosin portion of the actomyosin, that it was a phosphoprotein, and that actin reacted to displace the phosphate from the phosphomyosin intermediate. This chain of events is consistent with the picture of contraction of striated muscle obtained with the electron microscope.

### Synthesis of Protoheme

Protoheme is the complex of iron and protoporphyrin that, combined with particular proteins, is concerned variously in oxygen transport by red blood cells and in oxidations by tissues. The questions under consideration are whether protoheme is formed by addition of iron to the porphyrin or to some precursor, and whether the reaction is spontaneous or cell catalyzed. When extracts of duck erythrocytes, which have the capacity to form protoheme, are incubated with radioactive iron and the porphyrin, some of the iron can be recovered as hemin, a particular form of the heme. If either the porphyrin, the extract, or the incubation is omitted, little or no iron is recovered as hemin. This suggests that the porphyrin and the iron are the immediate precursors of the heme and that some component of the erythrocyte is necessary for heme function. Somewhat stronger evidence for heme formation has been obtained by more direct methods. Upon incubation of mixtures of

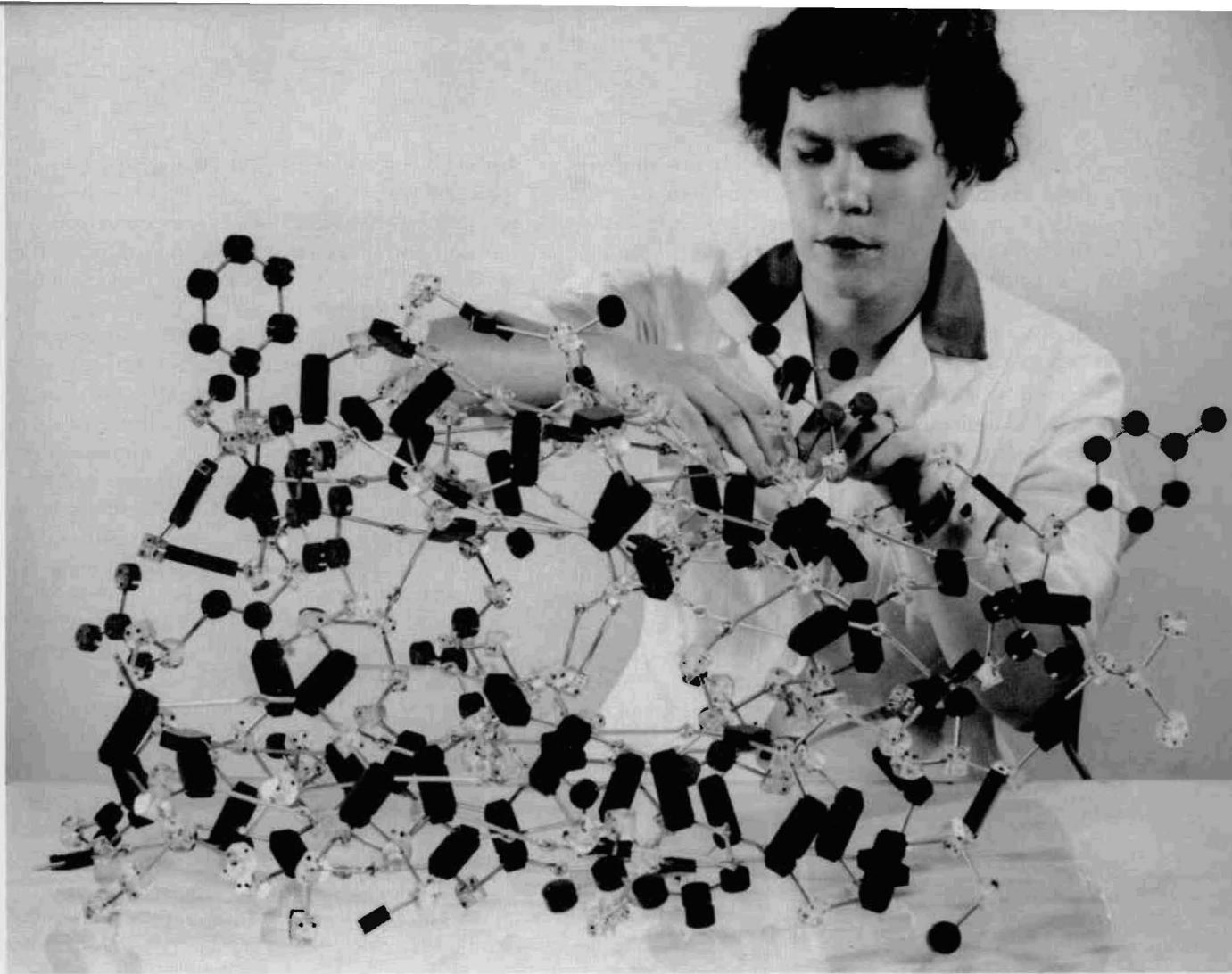


Figure 11. Molecular model of the portion of the protein phosphoglucomutase that is most profoundly implicated in enzyme activity. The model accounts for about  $\frac{1}{10}$  of the total amino acids making up a molecule of the enzyme. The small blocks represent atoms. The large blocks represent peptide linkages. The scale is about  $10^8$  times the actual size.

red cell extract, iron, and porphyrin, amounts of iron and porphyrin that are roughly equivalent to each other disappear, and heme in an equivalent amount appears. Again, the formation of heme depends upon the presence of the material from the cells. It is possible, although no evidence has been obtained, that under the conditions of the tests the porphyrin is converted to some earlier form that reacts with the iron, and the product is converted to heme. It is also possible that the material from the red cells simply provides a suitable medium in which the heme forms. However, at present it seems more likely that the porphyrin is the immediate precursor of the heme, and that the cellular material has an active function.

#### The Metabolism of Iron in the Duck

Study of the variations in metabolism among species has given insight not easily obtainable by study of any one species. Although the metabolism of iron has been studied extensively in mammals, particularly man, relatively little attention has

been paid to other vertebrates. In the present work the metabolism of iron was studied in the Pekin duck as reflected by the distribution within the animal of administered labeled iron. The duck was chosen as a representative of one nonmammalian species, experimentally convenient in many respects, and for which some pertinent information was available.

The labeled iron, given intravenously in amounts not exceeding 0.1 of the plasma iron, apparently mixed initially with about five times the amount of iron in plasma. The iron with which the labeled iron mixed conceivably was extracellular. In mammals, similarly administered iron is initially confined primarily within the plasma.

About 0.75 of the labeled iron appeared within the duck erythrocytes after two weeks or so. In man 0.90 or more of the administered iron appears in the red cells. The level of labeled iron in the red cells of the duck began to decrease after several weeks; from the duration of the decrease it appears that the red cells on the average live  $\approx 40$  days. The loss of labeled iron from the red cells was reflected by some increase in other tissues. This is taken to indicate that iron arising from the breakdown of aged red cells is handled in about the same way as is administered iron. Some of the labeled iron that entered skeletal muscle became a component of the pigment, cytochrome *c*, that is involved in oxidative metabolism. The rate at which the iron appeared as the pigment was slow compared to that of its entry into the muscle, and very slow compared to that of its appearance in red cells. In this respect the duck resembles the guinea pig and is in contrast to the rabbit. Some of the administered iron is excreted. The amount excreted corresponds to about  $10^{-3}$  of the total body iron per day, intermediate between the rates for man and the mouse. The excreted iron appears to be set aside for excretion some days before the excretion occurs.

### Metachromasy

When tissues are treated with certain basic aniline dyes for histological study, the color of the dyes changes in a characteristic way in the presence of particular tissue components. For example, cartilage is stained a pinkish hue by toluidine blue. This type of color change, which is unrelated to the color change of *pH* (acidity) indicators, is called metachromasy. The appearance or persistence of this metachromatic color under specific conditions has been regarded as a specific histochemical test for acid mucopolysaccharides. However, similar though less pronounced color changes also occur in simple aqueous solution when aggregation of the dye is induced. Consequently, a common explanation for metachromasy as it appears in the staining of tissue and in solution has been sought.

The dyes under consideration show marked absorption of light at two wavelengths. In the non-metachromatic state the absorption is greater at one wavelength than at the other. The appearance of metachromatic color coincides with depression of absorption at both wavelengths and a reversal of the relation between the degrees of absorption

at the two wavelengths. The change in absorption spectrum with appearance of metachromatic color is obtained when the dyes aggregate in aqueous solution and when bound to a suitable substrate, e. g., agar or appropriate tissue, or under suitable conditions the protein fibrin. Since the metachromatic color does not arise in the absence of water, approximation of the dye ions to each other does not alone account for the color. Also, the appearance of the color is not restricted to the presence of the acidic groups associated with mucopolysaccharides. The necessary conditions appear to be arrangement of binding sites, i.e., acidic groups, on the substrate for the basic dyes at intervals so spaced as to allow suitable approximation of dye ions to each other, with subsequent interaction, and interspersion of water. Thus, the pattern rather than the nature of the binding sites determines the color. The presence of closely spaced binding sites of one kind makes the acid mucopolysaccharides of special interest with respect to metachromatic staining.

## BIOPHYSICS

### Effects of Radiation on Enzymes

The composition, structure, and mechanism of function of enzymes, the catalysts of biological reactions, have been examined in a variety of ways. The use of radiation in this area of research, although comparatively recent, has provided considerable information. A problem of moment in the field of enzymology arises from the ability of some enzymes to act upon materials (substrates) of somewhat different composition. The question in such cases is whether the known interaction between substrates and enzyme occurs at more than one site on the enzyme. The enzyme under study, trypsin, which is involved in the hydrolysis of proteins in the intestine, can hydrolyze proteins, certain peptides (preliminary breakdown products of proteins), and also certain esters.

X-ray or ultraviolet irradiation of dilute aqueous solutions of trypsin inactivate the protease (protein-splitting) and esterase (ester-splitting) activities of the enzyme at different rates. This supports the impression that each substrate attaches to the enzyme at a somewhat different site. It is possible that the sites at which inactivation of the activities occurs are of different sizes (in the range of 100 to 200  $\text{A}^2$ ). However, this probably is not the explanation. Current experiments indi-

cate the presence in the irradiated solutions of at least three types of trypsin molecules: normal, damaged but still active, and inactivated. From the effects of temperature during the irradiation and the particular conditions prevailing in the three different methods used for assay of trypsin activity, it appears that damaged molecules can be converted to inactivated ones by rupture of four or fewer hydrogen bonds. The number of bonds involved may depend upon the activity assayed. The spectrum of radiation damage is not unexpected, since similar results have been obtained with the oxidants hydrogen peroxide and hydroxyl radicals, which are both implicated in radiation damage.

#### Properties of Films of Trypsin

Study of properties of films of biological material bears on the properties of surfaces or membranes at which many biological processes take place. Results from a study of the comparison of surface films of the enzyme trypsin and the recovery of enzyme activity have been used to evaluate four proposed structural models of the films. The film pressure,  $P$ , of the highly expanded (gaslike) films from which no activity could be recovered increased with concentration,  $C$ , according to  $dC/CdP = 1/P$ . Compression to concentrations greater than  $\approx 10 \mu\text{g}/100 \text{ cm}^2$  produces film pressure described by  $dC/CdP = \text{constant}$  (for the range  $2 \leq P \leq 16 \text{ dyne/cm}$ ). The value of the constant is a function of concentration and acidity ( $\text{pH}$ ). Films spread initially at higher concentrations also follow this relation; however, activity corresponding to the amount spread in excess of  $10 \mu\text{g}/100 \text{ cm}^2$  can be recovered. Compression beyond 16 dyne/cm produces a large increase in compressibility apparently attributable to crumpling of the skin. The findings indicate that some features of the four models considered are incorrect or improperly specified.

#### Delayed Radiation Effects in Seeds

When a biological material is subjected to an ionizing radiation, the effects of the radiation are usually considered to be dissipated within a small fraction of a second. These effects are produced by two mechanisms. The first involves direct ionization and consequent disruption of important molecules within the cell. The second is indirect and involves the disruption of small molecules, mostly

water, to form free radicals which then migrate to the important molecules and inactivate them by free radical reactions. The life of these free radicals is usually thought to be about  $10^{-9} \text{ sec}$ . Experiments with dormant seeds have shown that under special conditions the indirect effect of the radiation is not complete until hours or even weeks after completion of the irradiation. The implication is that the free radicals under these special conditions can have a very long life.

In these experiments seeds were irradiated and then kept for varying periods of time before being allowed to germinate. The growth at seven days as compared to that of the controls is taken as a measure of the damage produced by the radiation. This damage increases very rapidly for a few hours following irradiation and then continues at a slower rate for some weeks.

The effect of storage is greatly dependent on the moisture content of the seeds at the time of irradiation. Seeds having a very low moisture content exhibit the effect very markedly, especially those germinated after short storage periods. In seeds with a high moisture content this storage effect develops much more slowly and never reaches the magnitude attained in dry seeds.

The oxygen content of the water used to initiate germination is very important in determining the ultimate damage produced. If the oxygen content is high during the six hours immediately following irradiation, then the observed damage will be appreciably increased. After six hours the oxygen content of the water is unimportant. However, the damage continues to develop for many weeks.

These and other findings indicate that two processes are involved in the delayed damage. Both become immediately inoperative on the addition of water. One acts rapidly and requires oxygen to produce its effect. The other acts slowly and is independent of oxygen.

The storage effect is completely absent when the irradiation is performed with neutrons. This gives support to the idea that the storage effect is due entirely to the indirect effect of radiation; that is, the radiation acts on the water molecules, which in turn react with sensitive sites within the cell to produce the damage. With this interpretation it can be shown that under certain circumstances, for irradiation with x-rays, as much as 95% of the total damage is produced by indirect effects.

If the free radicals do indeed persist for long periods of time, they should be detectable by the

nuclear magnetic resonance method. Some very preliminary experiments with the magnetic resonance spectrometer seem to indicate that a resonance line corresponding to free radicals exists as

long as the storage effect continues. If this can be confirmed it will add great weight to the hypothesis that free radicals are causing the indirect and delayed damage in seeds.

## Medical Research

During Fiscal 1958 the research activities of the Medical Department continued to expand through intensification of investigations of many of its projects. The results of adhering to an integrated program of research for the Department were exceedingly gratifying in that close interdependence and interrelationship of investigations were obtained without sacrificing the individual scientist's independence in determining the phase of his investigation which appeared most productive for study. Observations made by colleagues not only frequently confirmed interpretations but also provided insight into situations which provoked the development of new hypotheses. Again the work in the Medical Department provides an excellent example of the interrelationship of fundamental research, applied research, and technological development as carried out by a group of skilled scientists within a broad area of a related group of scientific disciplines. It is clear that this report cannot include all details of the investigations but an attempt is made to point out the trends and significant observations which have been completed. The various headings under which the work is reported reflect certain operational groupings within the Medical Department, and the entire program could with complete justification be reported under an entirely different classification system.

### CANCER AND ALLIED DISEASES

Although the program of the Department is not primarily that of research on cancer, its problems are intimately related to the problems of cancer. This is necessarily so since the use of radioactive isotopes offers a most promising possibility for the treatment of cancerous conditions as yet beyond ordinary medical therapy. Furthermore, in attempts to utilize radioisotopes in this fashion, precise testing of our capacity to control the movement of the radioactive isotope within the body is carried out. This knowledge is of the utmost importance in the utilization of radioactive isotopes for general diagnostic procedures. Finally, since cancer may be provoked by radiation exposure, it

becomes not only of interest but of great moment to the users of radiation and radioactive isotopes to learn under what conditions and to what degree these types of tissue experiences may lead to malignancies.

The largest effort in the Department in point of view of people involved is the development of neutron capture therapy presently directed to the control of glioblastoma multiforme. During this year, the third series of ten patients received experimental treatments at the Brookhaven graphite reactor by a procedure sharply modified from that previously in use. In the present procedure the boron compound used was sodium pentaborate instead of sodium tetraborate and was administered by the internal carotid artery instead of via the antecubital vein. Two advantages were sought by this change in procedure, and it is believed both were attained. The vexing, exceedingly troublesome and serious skin lesions which were a complication in the second series of patients were avoided in the present studies. The response of patients who were deliberately selected from among those furthest advanced in their disease suggests that the retardation of tumor growth was greater than any previously attained. Until the pathological studies are completed these conclusions are only tentative surmises.

On other fronts, considerable progress was made on this project. It was definitely demonstrated by a complicated transplantation test following neutron capture therapy that a transplantable brain tumor could be rendered nonviable in something over 50 percent of the animals studied. Preliminary results utilizing the same treatment of this same tumor when an intramuscular transplant is used indicate that an even more satisfactory and complete result can be attained. Although in our patient therapy only the sodium pentaborate compound was used, test observations were continued with lithium and with uranium compounds. No data have yet come to hand that would suggest that either of these compounds is in any way superior to boron. We have continued to be concerned with the toxic effects of boron although these have been reasonably well

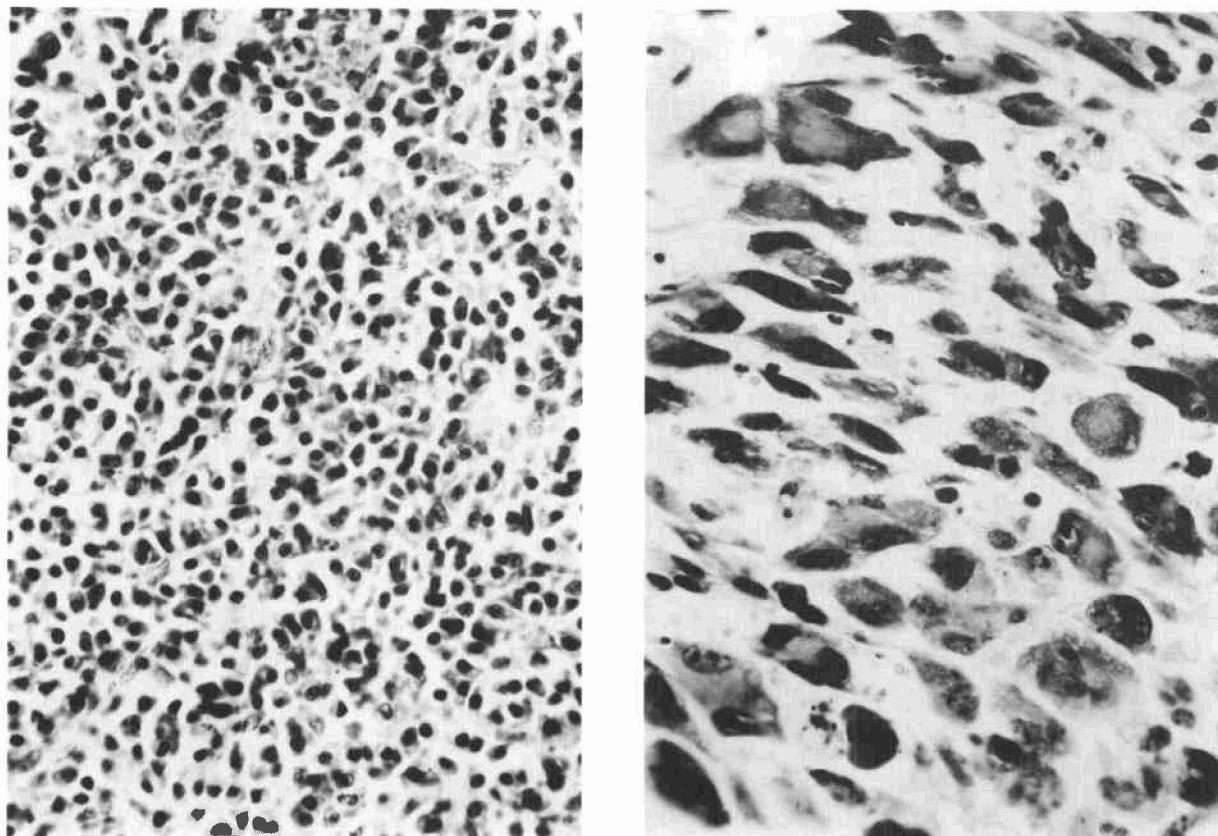


Figure 1. Histological studies of transplantable mouse sarcoma showing effects of neutron capture therapy four days after treatment. The section with the smaller cells shows essentially the usual tumor architecture; the section with the larger cells shows radiation effect. Both sections are at same magnification.

controlled, and a series of studies on the pharmacology of boron has continued in an effort to develop more effective control. Carbohydrates which may complex with borate ions were studied. The toxicity data obtained are currently being analyzed for lethal dose and relative potency value by means of the probit transformation maximum likelihood solution and relative potency techniques of Bliss and Finney. Since in this procedure the rate of transfer of the borate ions from the vascular system to cellular structures will be in part dependent on maintenance of vascular hemodynamics, numerous studies have been carried out to determine effects of saline and glucose solutions when given in large volumes. These latter studies will be correlated with boron distribution studies.

Further studies are proceeding on the distribution of both borax and sodium pentaborate in normal and neoplastic tissues in mice as a func-

tion of time. By this means it is hoped to determine the interrelationships of neutron exposure and boron atomic distribution for sure destruction of viable neoplastic cells while maintaining a high level of discrimination between normal and neoplastic structures. Of the boron compounds used, it appears that their distribution and concentration within the neoplasm and its several parts do not follow a smooth curve but proceed step-wise. The significance of this result is being investigated that we may be sure it is not an artifact.

In general studies on the efficacy of NCT an extensive investigation is being made on the various structures of the central nervous system for morphological effects of this procedure utilizing thermal neutrons. Serial sections of CNS obtained from patients at autopsy are being studied in collaboration with the Department of Neuroanatomy at Harvard and the Department of Neuropathology

at the Armed Forces Institute of Pathology. It has become necessary in this study to devote considerable effort to elucidation of the biology of the tumor, that the effects due to the disease *per se* and those due to the therapeutic manipulation may be clearly separated. The results of this study have been encouraging to date in that they indicate an absence of effects on normal structures and suggest some radiation effects on neoplastic structures. The desired and sought for chemical and biological discrimination can be rather clearly demonstrated in the mouse experiments, although as yet it is only suggested in patient studies.

The development of tritiated thymidine as a suitable label which can be demonstrated by proper radiographic techniques has had immediate application, particularly to the study of the genesis of the various cells of the blood. It has always been of importance to determine in leukemia whether or not an increased rate of synthesis or decreased rate of removal was the primary cause of the increase in cells in the peripheral blood. To a degree the same problem obtains with polycythemia vera. Utilizing this compound it was found that the labeling of neoplastic cells in the marrow of patients with multiple myeloma or chronic lymphatic leukemia was rare and that leukemic cells in blood of chronic lymphatic leukemia would not incorporate thymidine. Labeling was extensive in the marrow and peripheral blood of patients with acute and chronic myelogenous leukemia. The marrow of patients with infectious mononucleosis showed intensive labeling of primitive stem cells, and an increased number of labeled large mononuclear cells were present in the peripheral blood. The significance of these distinct differences in the different hematopoietic disorders studied and the potential value of these differences in establishing diagnosis, therapy, and/or prognosis are for further and continuing consideration.

With the demonstration that small doses of tritiated thymidine could be administered safely to patients, it was also possible to study patients with malignant diseases who had no disturbances in the hematopoietic system. The data obtained on these patients are similar to data obtained both on dogs and rodents. Within 15 minutes after intravenous injection of tritiated thymidine, labeling is largely completed. Labeling of large cells of coronal and central areas of the lymph follicles,

cortex of the thymus, white pulp and follicles of the spleen and undifferentiated cells of bone marrow is striking. For the purposes of study here, this group of cells has been termed the primitive proliferating pool. Serial marrow aspiration in the same patient and serial sacrifice in mice have shown a progressive diminution in the number of labeled primitive proliferating pool cells and a definite buildup in the labeled cells in erythrocytic, myelocytic, and megakaryocytic series suggesting, therefore, that the primitive proliferating pool cells represent a common progenitor pool for all lines of hematopoiesis. The degree of labeling in each cell is diminished only through mitosis. After four days, labeling in the bone marrow had largely disappeared which suggests many mitoses. The intense cellular proliferation of this large, widely distributed collection of primitive proliferating pool cells throughout the body was unexpected. The turnover rate of this group of mesenchymal cells may exceed that of any other tissue in the body. These observations may be interpreted as supporting the monophyletic concept of hematopoieses in adult life.

In another study, which during the past year has been to a considerable extent concerned with multiple myeloma but which has also included a variety of other malignant diseases, a detailed investigation of the physico-chemical nature of crystalline albumin and of gamma globulins and physiological studies on the rate of disappearance of these proteins labeled with radioactive isotopes have been carried out. The purpose of this latter procedure is to determine whether patients with various neoplastic diseases have similar or different rates of turnover of  $I^{131}$ -labeled protein derived from normal patients and of  $I^{131}$ -labeled proteins derived from cancer patients. Further experiments have been carried out to determine whether these same fractions have the same or different turnover rates in the rabbit. Certain differences have been observed in patients with multiple myeloma, the significance of which is still under study. During this past year a total of 30 patients were admitted for this project.

Studies previously carried out on the possible usefulness of  $Mn^{56}$  for the control and alleviation of metastatic malignancy in the liver were not continued in clinical phases during this year because it was not certain that the best method of administration has been developed. However, in-

tensive work on the metabolism of manganese has been carried out and will be detailed elsewhere in this report.

Previous work both here and elsewhere demonstrating that a significant single exposure to sub-lethal whole body radiation of the young Sprague-Dawley female rat is followed by the relatively rapid appearance of mammary neoplasms has been confirmed and extended. The incidence of breast neoplasia rises linearly with dose between 23 and 400 r of 250 KVP X-rays, measured at 11 months after exposure. Above dosages of 400 r the response does not increase. Ovariectomy preceding or following exposure to 400 r reduces but does not eliminate neoplasia of the breast. Intact males respond to the same degree as ovariectomized females, whereas castrate males give a response intermediate between that of intact males or castrate females and intact females. It would thus appear that the maximum induction of breast neoplasia following irradiation of these animals was affected by the level and type of gonadal hormones. Four distinct histological types of breast tumors have been observed - adenocarcinomata, adenofibromata, fibroadenomata, and fibrosarcomata. Mixed types are also seen. Shielding experiments have shown that neither the pituitary nor the ovary need be irradiated for induction of breast neoplasm. Results on effects following selective irradiation of the breast are not yet complete. Facts suggest that a single hit somatic mutation theory and an indirect or abscopal theory of induction of neoplasia are not necessarily mutually exclusive.

### DEGENERATIVE DISEASES

With the present control of infectious diseases achieved by the medical profession and the marked increase in our population, diseases of maturity have assumed perhaps the most important place in all medical programs. These diseases are largely intrinsic in origin, and many of them have been loosely grouped together as the degenerative diseases for purposes of convenience. In understanding these disorders which arise within the body and which are subtle in nature, prolonged in action, and destructive in result, tracer observations made possible with radioactive isotopes have the greatest promise for the logical development of procedures aimed at control of

these disorders. For as we know more about metabolism and its specific derangements, our experience has shown that we are better able to correct the derangements. During the past year patients with degenerative disorders have been studied here with diseases ranging from hypertension, on which a very extensive study has been carried out and is continuing, through arthritis, diabetes mellitus, and a variety of diseases of the central nervous system which tracer studies are increasingly suggesting may be associated with metabolic aberrations.

From many studies on hypertension there is considerable evidence that common sodium chloride plays an important role in provoking the disease. In salt it is the sodium component which is important. This, of course, suggests that some aberration of sodium metabolism either is associated with the disease or provokes it. The elucidation of this role of sodium is the subject of a continuing study in this Department. It has been shown in previous work here that the addition or subtraction of salt from the diet need not significantly change the sodium content of the body. A wide variety of other studies have been unable to indicate what the defective process is in patients with hypertension. It seemed logical, therefore, to focus attention on the blood vessels on the assumption that these are the primary site of the disease. Study of blood vessel physiology, however, poses severe technical problems which have not yet been solved. Therefore rat aorta preparations have been used thus far as an experimental analog, both to develop better procedures and to determine the nature of the processes in which sodium enters in the metabolism of our arterial cells. It has been found that differences in oxygen consumption occur in different regions of the aorta, and these can be correlated with regional frequency of atherosclerotic plaques in man. Since the blood vessels consist of smooth muscle and connective tissue, it is important to study both components.

The mucopolysaccharides in the intercellular matrices may be changed in their capacity to bind ions. Studies of collagen have yielded evidence of sodium gain and loss of such amounts that they cannot be explained by changes in the extracellular volume. These data suggest indeed that connective tissues in fact function as ion exchange systems and therefore may affect the smooth

muscle cells of the arteriole. A correlative to the project investigation of metabolism of blood vessels have been studies relating the intake of salt to hypertension in man. Studies done here indicate that the average intake of sodium chloride in Americans is about 10 grams per day. In lands or regions in which hypertension is said to be uncommon, a daily average of salt intake ranges from 1 to 5 grams. During the past year in connection with the Atomic Bomb Casualty Commission there was opportunity to study hypertension in a Japanese village of 1200 adults. Since the Japanese have a high salt intake, studies on vascular diseases carried an importance. In this over-all project, data are still being analyzed from this study.

#### Diseases of the Central Nervous System

In the study of degenerative diseases of the central nervous system, attention has primarily been paid to those disorders which have presumably been functional and morphological disturbances of the various nuclei and pathways. Some of these dysfunctions may be provoked by overexposure to metals under accidental conditions. For example, knowledge of the development of Parkinsonism following manganese intoxication led to the beginning of exhaustive studies on patients with Parkinson's disease for information as to whether or not these persons might distribute, absorb, excrete, or otherwise combine manganese in some unusual fashion associated with their disease symptomatology. During the past year this study has been expanded to include patients with Friedreich's ataxia and Wilson's disease as well as those with Parkinsonian syndrome.  $Mn^{56}$  has been the element primarily used because of its short half life and by virtue of its gamma emission permitting external scanning for body distribution. Patient studies have been strongly reinforced with experimental observations on animals. It appears, fortunately, that in many aspects the rat can be utilized to elucidate basic pathways of manganese in man. It is being increasingly suggested from the studies under way on patients that indeed differences in the handling of manganese may exist between the specifically diseased individuals observed and those who are free from this type of central nervous system disturbance. It is yet too early to hazard any prediction as to the precise nature of this aberration. Further testing and

specially designed observations are necessary to confirm the validity of the impression gained thus far. These will be continued in the year to come. It has been demonstrated that manganese pathways are exceedingly specific. Elements which are capable of displacing manganese *in vitro* have no effect on manganese *in vivo* as measured by tracer studies. Perhaps one limited exception to this may be technetium, and this point is under further study. Gallium, copper, mercury, zinc, and cadmium exhibited no capacity to displace manganese. Limited exchange occurred between zinc and cadmium which interestingly enough behaved in much the same fashion as the observed *in vivo* exchange between strontium and calcium.

As yet it has been impossible clearly to relate the complex capacity of phenobenzene drugs to their usefulness in controlling Parkinson's symptomatology. As this study has progressed year by year it has consistently revealed a considerable complexity of manganese metabolism which was impossible to demonstrate by other radioactive tracer techniques and which as yet cannot be satisfactorily fitted into a known pattern.

During the past year only a few patients with arthritis were studied. These individuals were given tracer amounts of  $C^{14}$ -labeled compounds to determine the incorporation of these compounds into orosomucoid. Other studies on orosomucoid are reported elsewhere as a part of the section on the charting of metabolic pathways.

The extensive studies in diabetes mellitus on carbohydrate metabolism and carbohydrate-fat interrelationship with  $C^{14}$ -labeled compounds were continued during this year. Observations were made on a number of patients to complete studies on the effects of prednisone on glucose synthesis and distribution. In diabetic subjects there is no evidence suggesting that the Krebs cycle and glycolytic pathways are different from those in nondiabetic individuals. The frequently low proportion of  $C^{14}$  observed in the outer carbons of glucose suggest that the activity of pentose pathways is minimal. In studies on the metabolism of  $C^{14}$ -labeled acetate and its conversion to glucose and cholesterol in patients, there was a moderate decrease in fatty acid synthesis from acetate in a stable diabetic patient as compared with that in a nondiabetic patient. This is in accord with various studies in animals. Young diabetics of either the juvenile or lipotrophic type showed greater

incorporation of C<sup>14</sup> in both fatty acids and cholesterol than stable diabetic or nondiabetic subjects. An unexplained effect of prednisone was a depression of 10 to 20 percent of the early appearance of C<sup>14</sup> in respired carbon dioxide from diabetic subjects. Analyses in one patient of arterial and venous carbon dioxide taken simultaneously with expired carbon dioxide showed a much slower rise in C<sup>14</sup> compounds in peripheral venous blood than might be expected, suggesting therefore acetate utilization by visceral rather than peripheral organs. Observations of urinary ketone bodies showed no change in C<sup>14</sup> incorporation after prednisone administration. Preliminary studies on distribution of C<sup>14</sup> in blood glucose after intravenous administration of C<sup>14</sup>-labeled pyruvate to diabetic subjects in part confirms previous findings from animal studies and is in part at variance from them. As expected from animal studies mevalonic acid has proven to be a very efficient precursor of cholesterol in humans. Extended studies of turnover rates of endogenous cholesterol were made. Four components of turnover were revealed with half lives of 99 days, 9.5 days, 2.7 days, and 10 hours.

#### **SELECTIVE KINETICS AND CHARTING OF METABOLIC PATHWAYS**

The utilization of radioactive isotopes for the elucidation of disease mechanisms, the specific diagnosis of diseases, or, in many instances, the treatment of a disease, requires exact and precise knowledge of the metabolic pathways of the element or compound used and a complete understanding and ability to predict the kinetics of distribution when specific placement and fixation or displacement and excretion are sought. It is clear that development of tracer theory is basic to these studies since it is not the behavior of the labeled material as such but that of the unlabeled counterpart which is usually of interest or significance. Therefore, it is highly desirable to establish the meaning of behavior of tracers in terms of events in the nonlabeled material. When, however, one is dealing with specific placement of a radioactive isotope for any precise therapy, the tracer itself becomes the substance of primary interest and import. In multicompartment exchange systems the relationships between the observed behavior of a radioactive isotope and the implied behavior

of the nonlabeled counterpart is complicated. In general, the interpretation and prediction of the behavior of the tracer rest upon a fairly elaborate mathematical basis. During the past year a review of the theory applicable to simple steady state closed systems was published. Extension of the mathematical basis of the theory of open systems and nonsteady state situations is being developed. The use of electronic analogs to simulate the biological situation and provide certain simplification of analyses of tracer data is being studied with a simple four compartment computer.

It will have been noted by the reader that in the application of radioactive isotopes, whether by the use of preformed radioactive materials or by neutron capture therapy, many projects of this Department are based largely on a chronological observation and prediction of radioactive isotope behavior, and these observations are heightened by selection of a radionuclide with an appropriate half life rather than by incorporation into organic carriers of known metabolic habitus. The complete understanding and charting of metabolic pathways are prerequisites for the most efficient utilization of chronological specificity. A very large number of metabolic pathways was studied by the scientists of the Department during the past year.

A considerable effort has been expended on the study of inorganic salts because of the presumed simplicity of their relationship to the systems involved. The studies are not bearing out this simplicity. Osseous mechanisms have been brought under scrutiny because of the necessity of understanding the factors affecting biological uptake and consequences of the deposition of nuclear debris in skeletal structures. A series of experiments were initiated with a view to elucidating the kinetic processes involved in the uptake and retention of minerals by bone and the relation of strontium-90 to these data. Analyses of the data have been performed using the theory of compartments. Resorption and excretion are considered separate compartments in these analyses.

In the study of rats, the accretion and exchange of the tracer were found to differ considerably among the various bones in the rats studied. The accretion rate per unit ash was highest in the ribs and lowest in the incisors. After 800 r of partial body irradiation, a decrease in the accretion rate was found. In contrast to this, the rate of turn-

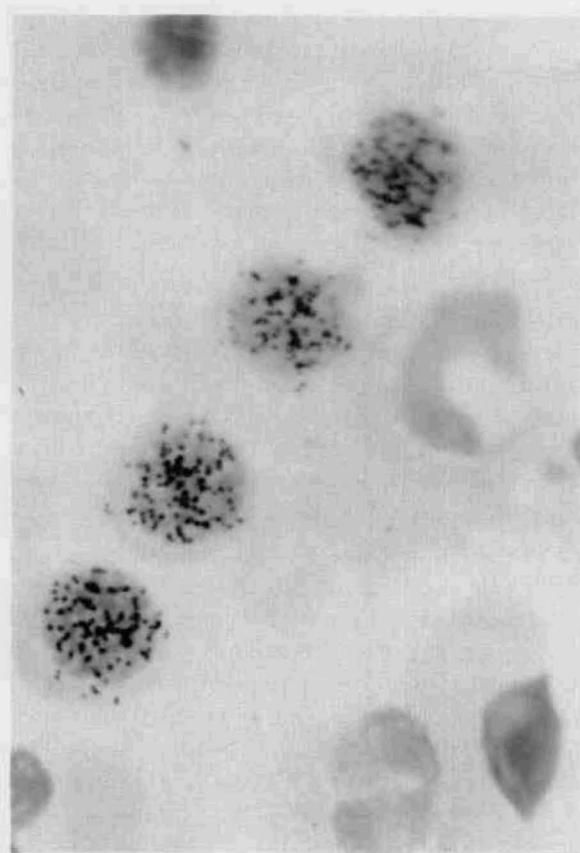


Figure 2. Labeled normoblasts in human bone marrow one hour after the intravenous injection of tritiated thymidine.

over of bone-fixed strontium was unaffected by 600 r. Further studies are in progress involving the use of  $\text{Na}^{24}$ ,  $\text{Zn}^{65}$ , and  $\text{Mn}^{28}$  for quantitating the rates for various metabolic processes in bone, particularly the surface limited rate processes.

As part of the general study of proteins, amino acids, and nitrogen metabolism, particular attention was given to nucleic acids and the synthesis of DNA by the use of tritium-labeled thymidine. Mice were given tritium-labeled thymidine intraperitoneally at various times following conception, and the uptake in embryonic tissue was determined. Significant uptake was not seen until the vascular system became established.

In another study utilizing tritiated thymidine, the proliferating capacity of rat organs was studied after the animal had received total body irradiation. Adult rats were irradiated with doses ranging from 500 r to 1500 r and were serially sacrificed from 1 to 72 hours and from 4 to 14 days.

Immediately before sacrifice, an intravenous injection of tritiated thymidine was given. Autoradiographs were carried out on autopsy materials from most organs. The injection of thymidine 30 minutes before sacrifice gives a measure of the actual proliferating capacity of all types of cell lineage, since thymidine is incorporated solely into DNA during the stage of DNA synthesis. The life of the animal subsequent to thymidine injection before sacrifice is designedly not sufficient for mitosis to occur. This study suggests that X-radiation in the doses given did not block DNA synthesis completely. In bone, the histological pictures point out clearly that regeneration starts from the endothelial cell layer of endosteum and sinusoids. These cells may be a part of the primitive proliferating pool previously mentioned.

Tritiated thymidine has also been given intravenously to selected human patients with clinically normal hematopoiesis. The results to date

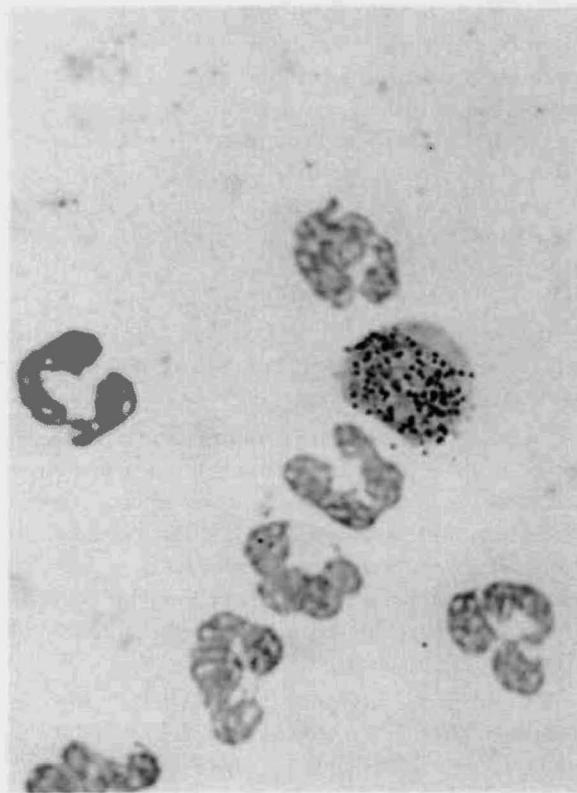


Figure 3. Large mononuclear cell in the circulating blood of a patient without hematological disorder, labeled by *in vitro* incubation with tritiated thymidine. Label indicates that the cell is synthesizing DNA, and thus presumably is capable of division.

indicate that tritiated thymidine leaves the plasma rapidly and its availability for labeling is measured in terms of a very few minutes. Subsequent to disappearance of thymidine from the plasma, from 30 to 60 percent of it is catabolized and tritiated water is excreted with a biological half life of 8 to 12 days. Very small amounts of nonvolatile activity are excreted in the urine a day after injection. There appears to be no significant amounts of tritium activity in the stool. From these studies it appears that about half of the administered thymidine is available for incorporation into body DNA. Clinical survey of the patients revealed no evidence of radiation effects in doses ranging from 3 to 19 mc.

Autoradiographs are being processed of specimens obtained from the skin of one patient taken at various times after injection of tritiated thymidine. Because of the continual renewal of the surface layers of the skin, it is believed this study may be of unusual interest.

All of the patient studies will be observed in the light of experiences gained in labeling cell nuclei of mouse tissues with tritiated thymidine. To date, autoradiographs show surprisingly large numbers of labeled nuclei which suggests that the amount of cellular proliferation in the mouse is greater than that inferred from a study of mitotic indices. The time for DNA synthesis appears to be a large fraction of the generation time of the intestinal mucosa. In the mouse 2000 r of X-rays inhibited cell production without markedly affecting the maturation or migration of cells into functional compartments.

In another study experiments were carried out using  $S^{35}O_4$  and tritiated thymidine. A study was made of the possible chemographic effects of tissue SH groups on radiographs. The level of penetrability of  $S^{35}$  and tritiated thymidine beta particles through celloidin coats to produce radiographs has been ascertained, and the findings reveal no SH grouping interference.

In additional investigations observing the metabolism of tritiated thymidine, the relationships of DNA synthesis time to mitosis and turnover time in HeLa cells have been studied. It was found that 20-minute exposure of HeLa cells resulted in excellent labeling of about 30 percent of the cells. From data obtained in this study, it was estimated that turnover time would be approximately the interval between the onset of labeled mitotic fig-

ures in the first cell division and the second cell division. These methods have shown that a culture of HeLa cells differs from its S3 clone in respect to DNA synthesis and mitosis. The serum component of the medium has also been shown to influence these parameters of DNA physiology in DNA culture. The effects of radiation on this system are now being analyzed.

As another tissue culture system, labeled kidney cells to be compared with HeLa cells have been used for tritiated thymidine uptake studies. Cells cultured for 24 hours in a medium containing 2.5  $\mu$ c of tritiated thymidine per milliliter showed heavy labeling, and there was no significant difference observed between uptake of tritiated thymidine by rabbit kidney cells and HeLa cells.

The use of tritium-labeled thymidine to mark cell nuclei led immediately to a need to know the concentration of thymidine that would inhibit cell division from radiation exposure to tritium. This is important in both the use of tritium-labeled thymidine as a tracer and its possible adaptation as a therapeutic material. Studies carried out showed an inhibition of growth when tritiated thymidine of high specific activity was used. HeLa cells cultured in a medium containing 1.5 to 0.5  $\mu$ c of tritiated thymidine per ml for a period of 24 hours contained large amounts of label. If the radioactive medium was then replaced by a medium containing unlabeled thymidine (10  $\mu$ g/ml) and cell death was observed microscopically over a period of 48 to 72 hours, counts showed that 40 to 80 percent of the cells died. Variation in cell death was noted to parallel rates in specific activity of tritiated thymidine in the cultured medium. Comparison with tritiated water showed that the same degree of growth inhibition was accomplished only when the tritium concentration was of the order of one to two thousand times that required with tritiated thymidine. These results were confirmed by a single cell plating technique. The cells which survived treatment resumed a normal growth pattern.

In a different type of study, labeled thymidine was used to mark DNA in order to permit observations of distribution of preformed and replicate nucleic acid components of bacterial cell nuclei during division. A culture of *E. coli* was used. The work obtained seems to demonstrate that the bacterial DNA is passed from parent to progeny by means of very large "pieces." Further work

has begun in order to define more precisely the number of these transferable units per cell. Studies on the effects of temperature and irradiation upon the distribution of label in progeny are not yet complete.

In another series of studies on the metabolism of nitrogen-containing compounds, extensive observations were made on the metabolic pathways of several amino acids and their derivatives or analogs. Because of its importance in metabolism, tryptophan has received a great deal of attention. It has been shown previously in this Department that anthranilic acid could partly replace tryptophan as a source of niacin, but it was not established that anthranilic acid was indeed an intermediate product in the formation of niacin. A method was devised for labeling anthranilic acid with C<sup>14</sup> in the carboxyl position. This compound has proved to be of considerable usefulness. Previous work in this laboratory has shown that 3-hydroxy-anthranilic acid appears in quinolinic acid, picolinic acid, and nicotinic acid. Recent evidence concerning anthranilic acid derivatives as antihypertensive agents increases the interest in this study.

In a complex and ingenious experiment, evidence was obtained that further suggests the formation of picolinic acid from tryptophan. In particular, the metabolism of C<sup>14</sup>-labeled kynurene and its conversion into nicotinic acid in the rat were studied, and attention was given to the questions: whether ortho-amino-acetophenone could function as a precursor to nicotinic acid or whether it could be oxidized to CO<sub>2</sub>. Observations were also made on anthranilic acid to determine whether the CO<sub>2</sub> was derived from the ring carboxyl carbon or both when the label was firmly established and whether it could function as a precursor of nicotinic acid. From studies in rats and particularly from evidence obtained from quinolinic acid and N'-methylnicotinamide, the data suggest that these products came from kynurene and that the carbon of the side chain carboxyl was retained in both products. It was also determined that ortho-amino-acetophenone is a structurally possible intermediate between kynurene and nicotinic acid. In the isolation of labeled quinolinic acid and N'-methylnicotinamide, the lower activity ratios of the products showed that the greater part of the excreted quinolinic acid and N'-methylnicotinamide was formed from sources other than the injected anthranilic acid.

In a study on species variation in the metabolism of 3-hydroxy-anthranilate to carboxylic acids by liver enzymes in which the cat and the rat were used, it was found that tryptophan labeled in certain positions led to labeled urinary quinolinic acid in both species but not to labeled picolinic acid. In both cats and rats, exogenous doses of picolinate were removed from the urine.

In another study of this system of metabolism, kynurene was labeled with tritium and it was shown that 70 percent of labeled tritium in kynurene was in the ring structures. This was administered to a rat and subsequently analyses of the urine were done. These showed that kynurene definitely is a step in the formation of niacin with quinolinic acid as an intermediate.

Amino acid metabolism was further studied using C<sup>14</sup>-labeled glycine to determine if it were utilized in ribose formation in regenerated rat liver. The data suggest a dual origin of ribose, partly from glucose by oxidative decarboxylation and partly by or from combination of 2 and 3 carbon units via transketolative reactions. The data suggest a difference of origin for ribose and deoxyribose.

Because of its wide occurrence among the proteins of the body, collagen occupies a unique place. Despite the fact that in such an animal as the rat it accounts for about 40 percent of protein, relatively little is known about its metabolism. Previous studies with C<sup>14</sup>-labeled lysine and hydroxylysine have shown that, in rats, the hydroxylysine is formed by the addition of an oxygen atom in the body and that there is no further quantitatively significant source of hydroxylysine in collagen. Even administered hydroxylysine is not incorporated into the collagen. The question of whether an amino acid can have its structure altered after incorporation into a body protein is significant in relation to the mechanisms of protein synthesis. Uniformly C<sup>14</sup>-labeled lysine was injected in rats, the skin collagen isolated and purified, and the C<sup>14</sup> specific activity of lysine and hydroxylysine determined. The results indicated that a rapid hydroxylation occurred during the first hour after injection of the lysine. It is surmised that this may have been at the time the lysine was incorporated into collagen. Subsequently there was a gradual decrease in the hydroxylysine activity ratio which suggests replacement of labeled contaminating protein by unlabeled protein. The proof of this sequence would be given by isolation

and identification of a contaminating protein or proteins, and this work is under way.

In a variant of studies on amino acid metabolism, the incorporation of  $C^{14}$ -labeled DL-tyrosine and DL-tryptophan by encysted *Trichinella spiralis* larvae has been carried out. The results show that the encysted larvae incorporate material fed to the host and that during the course of a *Trichinella spiralis* infection a significant change in tryptophan metabolism of the host appears to occur.

Studies carried out on cultures of the larvae showed that when serum was present in the culture media the larvae incorporated 35 percent of their total  $C^{14}$  as labeled tyrosine and tryptophan into protein but in serum-free media this percentage rose to 54 percent.

Another aspect of charting of metabolic pathways and the study of nitrogen metabolism is the work which has been going on on glycoproteins. Glycoproteins of human and bovine origin have been studied and appear useful for methodology and the development of certain comparative studies. In human alpha-1-glycoprotein, glucosamine is the only hexosamine found, but in bovine material galactosamine is found in an amount about one-seventh of the glucosamine preparation. The structure of these glycoproteins is important and it has been found that an enzyme in certain strains of *Clostridium perfringens* has the capacity to cleave sialic acid from orosomucoid and from several other sialic acid-containing proteins. Almost all of the sialic acid can be removed from the mucoid. A considerable amount of work has been done on this reaction because it is believed it will give insight into the structure of orosomucoid, the alpha-1-glycoprotein of human plasma. The technical aspects of these chemical studies are too complex for this report, but it may be pointed out that the sialic acid residues apparently occupy the terminal, nonreducing end of a carboxy chain. It would appear that the molecule probably consists of multiple small oligosaccharide units each linked to the polypeptide chain.

In another study using  $S^{35}$  on the mucopolysaccharide content of femoral periosteum and epiphyses of rats, it was found that very high levels of  $S^{35}O_4$  uptake at the periosteum occur at birth. After eight weeks and a diminution in uptake, the avidity of this structure for sulphate became more constant. It is believed that the acid mucopolysaccharide found in the elements of periosteum is as-

sociated with the mitochondrial complement of the periosteal cells, especially the osteoblasts.

The degradation of gamma-2-globulin has been followed in mice. The material was iodinated with  $I^{131}$  after isolation from the blood of normal persons and also from the blood of patients with multiple myeloma. At the present time it appears that the half life in mice of gamma-2-globulin from multiple myeloma patients is not different from the half life of gamma-2-globulin from normal persons.

Although a major part of our program in chemistry and metabolism is not devoted to the study of lipids, it is obvious that one cannot consider the metabolism of proteins and carbohydrates without from time to time carrying out studies on lipid metabolism since so many interactions occur. During the past year several lipid studies were done. The *in vitro* synthesis of lipids by human red cells was observed after incubation of 50 ml of whole blood with a  $C^{14}$ -labeled sodium acetate under appropriate conditions of  $pH$  and stability. It was found that in plasma, free cholesterol was labeled while the esterified sterol had no radioactivity. Free fatty acids in plasma were highly labeled while triglycerides exhibited negligible incorporation. In the red cells, the activity of triglycerides was quite high, being seven times that of the free fatty acids which were only one-ninth as radioactive as the plasma fatty acids. Red cell esterified cholesterol showed an unexpected high specific relative activity. The phosphatide fatty acids of both red cells and plasma had nearly equal activity.

In another study, carbon-labeled mevalonic acid and tritiated water were injected intravenously into a growing pig to determine incorporation into cholesterol of blood, brain, liver, and muscle. Analyses with specimens obtained in this study have not yet been completed, but it is hoped to obtain information on the synthesis and exchange of cholesterol in these organs. When swine are fed a diet deficient in essential fatty acids, they develop a disease called Parakeratosis. From the data presently at hand, it would appear that the turnover rates of plasma lipids in the normal and the mildly diseased pig are essentially the same.

Since there is considerable interest in the Department in the central nervous system, it might be expected that studies would be carried out dealing with carbohydrate metabolism of the central system. Utilization of  $C^{14}$ -tagged sugars is useful in studying the problem of transfer of sugars

passing the blood brain barrier. Cats have been used in this study. It was found necessary to make further observations of the carotid circulation in cats because of numerous abnormalities which might seriously affect the integrity of the interpretation of the results. Otherwise what might be believed to be a contained cerebral circulation might have adventitious sources. A study utilizing these sugars has sought to determine the systematic specificity of the monosaccharide permeation carrier system in the human red cell. In this study it is suggested that preferential reaction is correlated with increasing stability of the sugars in the pyranose C-1 chair conformation. This type of steric consideration has not previously been implicated in connection with biological specificities. Studies of inhibition of this system by hydrophenyl substances have been carried out. The close parallelism noted previously between the sensitivity of the red cell system and the oxygen consumption of mouse Ehrlich ascites tumor cells has not in every instance held true in all these analogs.  $C^{14}$ -labeled glucose tracer studies appear to offer new and independent evidence of the validity of the carrier hypotheses for glucose transfer within the red cell. In the brains of cats, isolated cerebral circulation perfusion studies have been carried out successfully *in situ*. Electroencephalographic tracings remain relatively normal and the peripheral nervous connections were intact. Because of the complexity of the preparation used, only a very few observations have thus far been made on the effects of extracerebral factors in the transfer of sugars between blood and brain. It is believed now, however, that this preparation will be useful in determining the significance of conformational variations in sugar structures *vis-à-vis* their ease of transfer to the central nervous system.

In the studies concerned with charting of metabolic pathways, the inorganic salts were originally assumed to be simple and the pattern of movement known. Increasingly detailed observations of various inorganic salts, however, have indicated an unusual complexity in the metabolic pathways followed by these crystalloids. A very considerable attention has been directed to the charting of the metabolic pathways of manganese. As previously noted, this has both therapeutic and diagnostic implications. Since manganese is excreted via the bile, it was obvious that more knowledge regarding this mechanism was desirable. In the bile, manganese becomes 10 times more concen-

trated than it is in the blood during any one time period. In albino rats a maximum excretion rate in the bile is noted at 60 to 90 minutes. This maximum is intensified by intraportal injection of the isotope and is not abolished by complexing manganese with versene or by poisoning the animal with dinitrophenol. It is specific for manganese and is independent of total bile flow. A second component, evident in the bile, disappears in conditions of manganese deficiency and can be seen only in animals on a high manganese intake. Mention has already been made of the high specific pathways followed by manganese. Elution from the body of radioactive manganese cannot be accomplished with magnesium, transition group metals, or members of the 7th group of elements. In this regard, it is quite different from nearly all other metallic ions studied. For instance, studies have been made of the replacement of bromide by chloride, molybdenum by tungsten, strontium by calcium, and to some extent zinc by cadmium. It is not known whether this high specificity of manganese is entirely due to the compound with which it might complex or to the specific loci within the cells where it may largely be found. Manganese appears to follow somewhat different pathways depending upon its route of injection and its metabolic experiences prior to the time of observation. The significance of these diverse routes is not at the present time clear. For instance, in general the transfer of manganese from blood into the intracellular sites was rapid throughout the body. Nearly all endogenous manganese is available for exchange with the tracer within an hour. Yet manganese distribution is in three and possibly four body compartments. Data suggest that mitochondria may constitute one of these compartments and that manganese kinetics may provide a method for measuring mitochondrial mass in intact mammalian bodies. Although manganese appears to behave in a highly specific way, no differences could be demonstrated, insofar as exchange of body manganese with radiomanganese was concerned, when different forms of manganese were injected. Chromous and ferrous citrates did not cause any deviation from normal partition of manganese in the body. It seems quite likely that unidentified organic compounds may play a significant role in delimiting manganese specific pathways. Manganese excreted is entirely bile secreted whereas for other elements the real excretory route is primary. At the present time, it is

not clear whether or not this is significant in relation to manganese.

Among the systems of the body which have been observed specifically and intensely none has received more attention in this Laboratory than immune mechanisms. On the one hand, immune mechanisms can be considered as a special illustration of protein metabolism and on the other hand as a specific response of a given type of tissue to certain stimuli. Not only are the specific mechanisms of immediate interest but also the utilization of specific locations and of migration of body elements and their temporal relationship. The exaggeration of anaphylaxis while antitoxin production is suppressed after radiation raises complex and interesting questions regarding the composition of components of immune systems. In an attempt to determine certain cell types which may be involved, a correlation was attempted of tetanus antitoxin formation with the histology of intraocular transplants of thymus. The data thus far suggest that the immature cells present in this transplant two to ten days after transplantation are the cells which then and latterly have the ability to respond to the specific antigen and produce tetanus antitoxin. However, the participation of other cell types cannot be excluded at this time.

When the secondary tetanus response in mice was studied it was demonstrated that doses of 48 to 288 rep given at a dose rate of 4 rep per hr depressed antitoxin formation. Comparable doses of this amount of acute gamma radiation did not similarly depress antitoxin production. Higher acute doses did, however, and with much larger acute doses, an accumulated dose of about 2700 rep was needed to depress antitoxin formation to the level observed after an acute dose of 650 rep. These differences in response to radiation are being studied further.

Attention was devoted to degradation of antigens in a study of metabolic pathways. The use of labeled proteins and whole body counting provides new techniques for determining the rate of degradation of protein antigen. The study concerned itself with the quantitative relationship between the amount of antibody used in the sensitization and the amount of antigen degraded at an accelerated rate. When the amount of antigen degraded at an accelerated rate was determined, it was found that the maximum occurred when there was a significant excess of antigen and when conditions were such that little if any antigen was precipi-

tated *in vitro*. Preliminary experiments with horse and mouse antisera indicate that variations exist between them which may be based on qualitative differences.

Another facet of the degradation of antibody was the study in mice of acquired partial tolerance to crystalline bovine serum albumin. The antigen was given both in single and in repetitive doses to animals from birth to four weeks of age, and the existence of immune tolerances was tested by attempted sensitization for fatal anaphylaxis. Mice given four injections begun at birth or one week of age demonstrated complete tolerance at six weeks of age. The degree of tolerance decreased with time. Single exposure was not as effective as multiple exposure in inducing immunological tolerances. The tolerance state is transient and since it is frequently followed by active sensitivity it is apparent that there is only a suppression of immune mechanisms and not an elimination of them.

Previous mention was made of increase of susceptibility to anaphylactic shock following irradiation. When a continuous exposure to gamma radiation at a relatively low rate was given to a total of 192 to 288 rep, a sharp increase in susceptibility to fatal anaphylaxis was observed. Recovery from this enhancing effect began during the second week and was complete during the third week. The antihistaminic agent Thephorin (R) afforded complete protection from this fatal anaphylaxis.

The importance of knowing the time relationship of boron distribution required numerous additional studies during the past year. The temporal changes of boron concentration in peripheral and central tumor areas were contrasted as well as between proximal and distal areas wherein the direction was along the course of the vascular stem. Consistent differences were noted in these various tumor regions of the transplanted mouse tumor. Observations were also made of the time of appearance of boron in the skin of mice and of patients after intravenous injection. In the pig, a study was done to determine in part the genesis of the skin lesions noted in patients. It was conclusively shown that this is the result of a capture reaction and could be avoided by utilizing irradiation times when boron concentration was at a minimum.

In other studies on the metabolism of amino acids, a leucine-requiring mutant of *E. coli* was used in the belief that it would be useful in de-

termining the pattern of protein partition during growth after labeling with tritiated leucine in a period when DNA synthesis is known to be occurring. At the present time data do not reveal whether the pattern of distribution of nucleoprotein, among progeny in this instance, is merely one of dilution or one of a more complex nature.

Among other studies on the metabolism of nitrogen-containing compounds has been one on the catabolism of serum containing albumin when the albumin has been labeled with  $I^{131}$ . It was desired to determine which organ or organs might be concerned in the metabolism of this compound. Partial hepatectomy in rats markedly decreased the rate of catabolism, whereas removal of the intestines appeared in fact to accelerate catabolism. Reticulo-endothelial blockade decreased the catabolic rate. These results combined with those of previous studies suggest that the serum albumin was catabolized after diffusion across a semipermeable membrane. It is of interest that the kidney may play a significant role in the catabolism of this protein.

### RADIATION EFFECTS

The effects of radiation are a primary concern to all members of the Department. Many radiation effects are inherent in the observations which are made for other purposes, and studies of this type will therefore not be repeated under this section.

Studies on the morphological effects of radiation include observations wherein the radiation is provided by: energetic lithium and alpha particles resulting from thermal neutron capture in boron-10, kilovolt protons from an accelerator, and beta particles of various energies as from manganese-56 to tritium. It can be seen that these types of radiation provide experiences ranging from a molecular to a regional irradiation. Gamma sources and X-rays are also used to complete our experiences. Not only are we interested in the immediate effects of radiation but also in the late effects of radiation. One of the areas in which we have a very high degree of interest is what might be termed validation of tolerance doses, i.e., an evaluation wherein it can be definitely stated that a summation of chemical, physical, and physiological examinations to determine the dose received did not demonstrably alter the system of interest and that therefore, within the limits of the study, the tolerance dose

was validated. It is apparent that radioactive isotopes can be used successfully as tracer materials if the incorporation of an energetic atom in a large organic molecule does not result in physiological instability.

Relative permanence of the tracer atom within the nuclear structure, stability of the final molecular compound, and some degree of equilibration, attachment, or placement within the body must be accomplished in most instances before any radiation effects can be evaluated.

As part of our study on radiation effects there is under way an animal project on the therapy of the hematopoietic syndrome of acute whole body radiation injury. In one phase of this study 20 dogs received 400 r of X-radiation; half of the irradiated animals were treated and half remained as a comparison group. Nine of ten animals in the comparison group died while eight of ten in the therapeutic group survived. Experimental therapy consisted of antibiotics, fresh whole blood transfusions, parenteral fluid infusions, forced oral feeding, and meticulous nursing care. The antibiotics were given as necessary to control infection. In the treated dogs that survived, the platelets, leucocytes, and red blood cells did not return to their pre-irradiation values until 60 to 90 days following irradiation. This therapeutic regimen significantly decreased the mortality rate in dogs. In the present study, this regimen will be explored at even higher doses.

In another study in animals rendered thrombocytopenic by whole body irradiation, a semi-quantitative measure of the degree of bleeding was obtained by enumeration of red blood cells in thoracic duct lymph. After exposure, large numbers of rbc were observed in the lymph. Lyophilized dog platelets did not increase the recipient dogs' circulating platelets, nor did the number of rbc following its administration in the lymph decrease significantly. When fresh platelet preparation was given, circulating platelet levels increased and there was a drastic decrease in the red blood cells of the lymph. Similar observations have previously been made on patients in whom freshly prepared platelet suspensions were injected.

The hematologic effects of gallium-72 administered intravenously to patients with diffuse bone metastases or primary bone malignancies were studied. The effects on this system of this internally administered isotope were in general

similar to those results seen in whole body external irradiation. It would appear that the depth and duration of platelet count depression are the best index of the degree of marrow damage after irradiation, and they might be indicative also of the total damage sustained by the marrow from previous exposure. A total white count below 1000 and a platelet count below 25,000 may be tolerated for weeks without infection or gross bleeding and with ultimate recovery. Hematological therapy because of these findings alone is not indicated. It is clear that the significance of hematological depression both as a measure of therapeutic effectiveness of radionuclides and as a measure of absorbed dose requires close observation of all patients receiving isotopes.

One area in which uncertainty continues is the concept that when the radiation dose is expressed in units of absorbed energy only, such as rad, various types of radiation require different doses to produce the same kind and degree of biological effects. This has been termed the relative biological effectiveness and is currently referred to as rbe. Explanation of this phenomenon in terms of other physical parameters such as linear energy transfer has not been entirely satisfactory. To investigate the rbe of heavily ionizing particles in the mammalian system, the mortality rate of mice injected with  $B^{10}$ -enriched borax and irradiated with thermal neutrons was studied over a large range of nvt and mean tissue boron concentration. Results indicate that the degree of effects varies with the product of the nvt and boron concentration.

Some work has been addressed to the theoretical and practical question of whether previous exposure to ionizing radiation can induce any degree of resistance to a second exposure. Goldfish are being used for the experimental animal, and an initial exposure of 200 r is being used. Thus far the experiments to determine if the LD50 value as well as the survival time has been altered have not been conclusive.

### ACTIVATION ANALYSIS

The utilization of selective activation of elements for analytical purposes, particularly for use with biological specimens, is being developed as a separate project for general application. It is perfectly clear that one type of activation which is being widely exploited in the Medical Department is neutron capture therapy. In the present

study the prime goal is that of determining trace metal concentration in biological samples as well as that of measuring other elements which may occur in significant amounts in tissue specimens. The work revolves about two general phases: one is the activation and the other is the analysis. In both instances collaboration is being carried out with other departments, namely the Physics Department, the Reactor Division, and the Department of Instrumentation and Health Physics. It is believed that through the use of proper filters and screens and with proper coincidence circuits a highly effective system of analysis can be developed by means of these principles. As yet this system has not reached the stage of any application, although some preliminary trials have been carried out on the analyses of electrolyte content of specimens obtained from arterioles and large arteries in the rat.

### INSTRUMENTATION AND DEVICE DEVELOPMENT

In the application of numerous instruments, it is necessary to adapt them for the specific study which is being carried out. In other instances, devices must be developed which are capable of doing the task required. Various types of instrumentation have been developed, and there has been a considerable amount of device development during the past year. In the Brookhaven research reactor and at the Materials Testing Reactor, targets consisting of 100 mg each of highly enriched  $Sm^{144}$  were activated. These sources were then inserted into a device designed for taking radiographs with thulium-170. The device was loaned to Brookhaven by the Army's Instrument Development Branch at Fort Totten, N.Y. Radiographs were made of nonliving objects and of a few human extremities. The results show that radiographs suitable for the diagnosis of fractures can be obtained with  $Eu^{155}$  as the source. The definition is good but the contrast and therefore the detail of bone structure are only fair. A number of other targets have been activated for study of methods for separating  $Eu^{155}$  quantitatively from the samarium target. A method involving selective elution from an exchange column shows promise of being adaptable to repetitive activations of a given target. The chemical separation procedures are being studied further for application to targets sufficiently large to provide the multi-

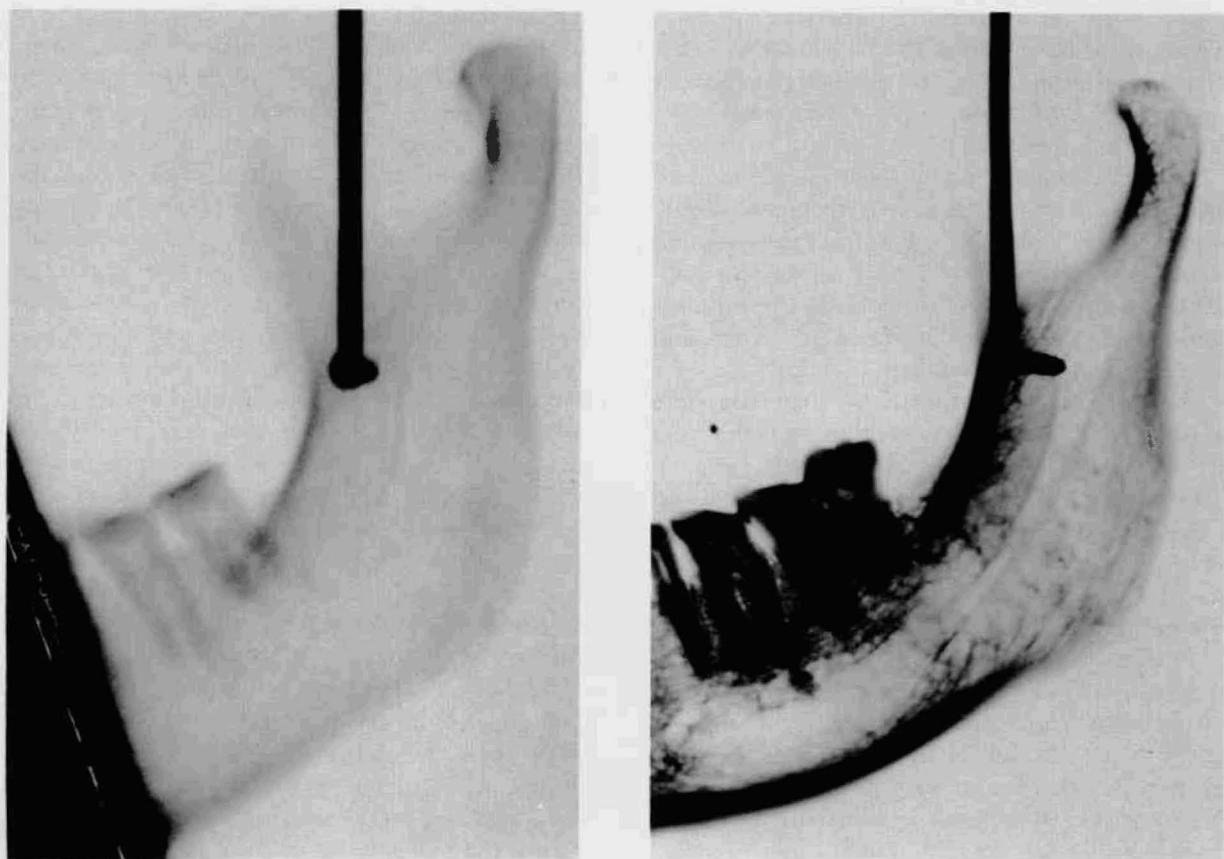


Figure 4. Comparison of radiographs of human mandible taken with europium-155 (left) and with 50-kv X-rays (right).

curie strength required by Eu<sup>155</sup> sources to reduce exposure times to convenient intervals.

In studies at the reactor and in the administration of energetic radioisotopes, it is frequently necessary to be able to administer material at a constant predetermined rate which may be changed with time and to be able to control the device at some point distant from the injection site. Most pressing need for such a device occurred at the reactor during neutron capture therapy where it was necessary to begin the infusion with glucose but at a predetermined time to change the infusion to a boron compound and to administer this at a constant rate which usually was different from the rate of administration of glucose. In conjunction with members of the staff of the Hot Laboratory, a very satisfactory apparatus was developed which provided for administration of either of two solutions. The containers for these solutions were connected by a specially designed three-way

electrically operated stopcock connected to a Sigma motor pump. Water of constant temperature was circulated through jackets surrounding the reservoirs of infusion fluid and drug-carrying tubes. All units were capable of effective sterilization. This device proved to be exceedingly useful in carrying out injection for neutron capture therapy and it holds promise of use for the injection of very active radioactive isotope solutions.

The large variety of isotopes and types of radiation which have been used in the Medical Department have posed numerous problems in the field of radioisotope assay. It is desirable to count energetic gamma emitters *in vivo*, whereas soft  $\beta^-$  emitters such as tritium require methods that involve putting the active sample inside the detector system. A beta-ray source comparator was developed for precision and reference measurements in the standardization of isotopic sources. It is used to evaluate flat surface applicators and pre-

pared aliquot standards of radioactive solutions. The apparatus is based on the Failla extrapolation chamber design; the rate-of-drift measurement principle is employed with a Lindemann electrometer as indicator.

A commercial *in vivo* radioisotope scanning unit was found to be inadequate both mechanically and electronically and is being entirely rebuilt. Another commercial device for radioisotope uptake measurements was also found to be unstable and was replaced by equipment designed and constructed by the Instrumentation Division.

Several special counters have been put into service for simplified measurement of radioactivity in small animals and in excretion samples. These include a large well scintillation crystal and a geiger counter assembly of sufficient size to surround a standard daily urine collection bottle. Counting with these devices is done with simple discriminator limiting circuits.

Proper application of massive neutron fluxes in medical and biological investigation calls for detailed knowledge of the quantity and quality (or energy) of the available neutrons. An adequate estimate of other radiations which may be present is also needed. Reactor slow neutron experiments have relied on foil activation as a simple standard detector using gold in the form of thin sheets or wires; cadmium ratios have been used for a rough measure of energy spread. The adaptation of more refined techniques has been limited by space and time requirements in both reactor and intermediate energy accelerator neutron researches. Distribution patterns for neutron-induced activities were explored in solid and in liquid tissue-equivalent phantoms. Similar instrumentation and phantoms were used in the Civil Effects Test Program at the Nevada Proving Grounds.

While the neutron capture therapy program has continued, including the ancillary investigations with small animals, the recent refueling of the main research reactor has resulted in several serious changes of the radiation pattern available from the medical facility. Of especial importance is the unexpected reduction in neutron flux, attributable to the narrower uranium loading pattern as well as the consequent shift of large neutron sink targets into closer proximity with the medical exposure port. Another factor of great importance is the alteration of the neutron energy spectrum; this has not yet been evaluated in detail since the simplified foil activation system used for standard-

ization of medical exposures is not versatile enough to indicate the changes. Additional methods are being applied to establish these factors.

Neutrons of intermediate energies are being investigated for their relative radiation effects. Physical and biological dosimetry has been developed to evaluate neutron efficiencies through the energy range from 0.1 to 2.0 Mev. Tissue-equivalent ionization chambers of the condenser type are used for the physical measurements. The end point for the biological evaluation in mice is testis weight loss. The initial work utilized neutrons from the proton - tritium reaction in the 18-in. cyclotron. Further work is planned with this machine when it is returned to service; meanwhile additional pilot experiments are scheduled for the Van de Graaff accelerator. This program is in cooperation with the Physics Department and with the Radiological Research Laboratory of the Columbia University College of Physicians and Surgeons.

Work on the medical research reactor has been going rather slowly during the past year because of delays in completion of the building. As of June 30th, construction of the housing and containment shell was not even yet completed. On the other hand, the component parts of the reactor itself have been delivered and constituent units of the control system, graphite reflector, and other segments have been received and assembled for checking and for operational testing.

At the same time explorations have been carried out on the design of another type of reactor which might be useful in solving physiological problems of immediate moment. These problems are at present not susceptible to attack with the reactor which we expect will become operative during the coming year.

## METHODOLOGY

In carrying out any research program it is constantly necessary to devise new methods and to improve old. In some instances the methods are concerned with the establishment of norms instead of specific analytical procedures. When specific effects of a procedure may be known, it is then possible to determine the experimental variance in which primary interest may lie. Such a problem deals with the effects of phlebotomies on the platelet levels with polycythemia vera. Since we are interested primarily in platelets, it is necessary

to know what ancillary procedures may do to platelets if one is to control diseases with radiation or with radioactive isotopes which also have an effect on platelets. In this instance a study of the platelet counts of patients done serially before and after venesection and with  $P^{32}$  and iron is under way. To date the usual thrombocytosis seen in normal individuals following a large venesection has not been observed. These clinical analyses of a method are continuing.

A standard apparatus originally devised by Van Slyke and Neill for blood gases has found application in other fields, including that of radiochemistry, e.g., in Hugo Fricke's studies of products formed under the influence of X-rays, and in the Brookhaven studies of the reactor gases and measurement of the occluded nitrogen gas formed in sodium azide under the influence of neutron beams, and as part of the equipment for determining organic  $C^{14}$ . A micro form of the extraction chamber has been devised and tested in which the extracted gases are brought to a volume of 100  $mm^3$  and measured by the pressure they exert at that volume with a standard error within  $\pm 0.25$  percent. To keep the precision of sample measurements within similar limits, special pipettes have been devised for measuring liquid samples of 50 to 100  $mm^3$  volume into the extraction chamber. The micro apparatus has been used in clinical blood analyses, and also in measuring microgram amounts of  $C^{14}O_2$  preliminary to counting in the Bernstein - Ballantine gas counter.

In order to use tissue cultures, it is necessary to establish satisfactory procedures for their propagation and to become thoroughly familiar with the changes in the cells that may be seen under such a so-called standard environment. During the past year, collaborative studies with the Viral and Rickettsial Research Division of Lederle Laboratories utilizing a cell culture of adult rabbit kidney cells which was established in our laboratory since 1956 were carried out. Careful consideration was given other culture requirements of rabbit kidney cells. By an analysis of nutritional needs, it was possible to develop a medium permitting the growth of single cells into microscopically visible colonies consisting of large numbers of cells. This made it possible to clone the original culture and establish an homologous cell population.

In biological chemical analysis it frequently is exceedingly effective to utilize an enzyme which

will cleave a compound of interest from a complex parent compound. It was previously reported from this laboratory that an enzyme is present in culture filtrates of *Clostridium perfringens* which cleaves sialic acid from orosomucoid. The conditions under which the organism produces useful levels of activity have been studied. The results suggest that the enzyme is an adaptive one rather than a constitutive one. It is now possible to grow *Clostridium perfringens* under conditions which yield predictable levels of enzyme activity. A gram negative organism has been found which appears to require orosomucoid for its growth, and the bacterial yield of this organism has been shown to be dependent upon the concentration of the orosomucoid in the medium. At the present time, studies are under way to determine the mode of utilization of substrate that there may be further development of this capability into biological method.

The utilization of the circulatory system to deliver radioactive isotopes to malignant structures highlights the question which has long been under consideration of the effective physiological development of vascular structures of neoplastic tissues. A method has been under study utilizing radioactive cobalt and radioactive molybdenum labels for red blood cells; by this means a measure of the physiological vascular volume in a tumor of known weight could be determined. In sarcoma transplanted to mouse brain, it has been shown that there are at least three times as many circulating red cells as in the same weight of normal brain. This is not believed to be due to stasis or hemorrhage. A satisfactory method of determining the plasma volume in neoplastic tissue has not been evolved.

To facilitate calculation of dose rate after internal administration of radioactive isotopes, a nomogram covering all isotopes presently in use in medicine and biology was constructed.

During the year further studies have been carried out on the development of methods utilizing light scattering for the characterization of molecules, particularly proteins. It is hoped that this method may ultimately prove useful in yielding information on radiation effects on soluble proteins.

During the past year further steps were taken toward developing our program on low level whole body counting. This was markedly accelerated by the need of a whole body counter for use in the project carried out under the auspices of

this Department in the Pacific area. A unit was designed and constructed which was transported to the Marshall Islands and proved to be effective in studies directed at determining the nature and quantity of radionuclides internally deposited in the residents studied in the Islands. The experiences gained are being applied in the design and construction of the whole body counter to be installed in our new building.

During the year the Department continued to carry out a number of special projects most of which are continuing but with significant variations during each reporting period.

The Department continued its responsible direction of the medical studies of the Marshallese who were subjected to radioactive fallout in March 1954. An 18-man medical team composed of research and technical collaborators of this Department was transported to Rongelap for the examinations in March 1958. Complete history

and physical examinations including growth and development studies in the children, hematological examinations, ova and parasite survey, and whole body gamma spectroscopy for body burden of isotopes were carried out. The expedition, having satisfactorily completed its task, returned to the U.S. leaving equipment and data to follow at a later time. Because of engine trouble on a plane transporting equipment and some of the data, it was necessary for the pilot to order all cargo jettisoned. The most important data lost were the gamma spectroscopy measurements. Consequently extraordinary efforts were required to reassemble equipment, and in May a small team returned to Rongelap and repeated the whole body gamma counts. In the years to come further emphasis will be placed in these surveys on phenomena of age and disease incidence which may be related to the radiation experiences of the people. To many members of the Department



Figure 5. Varied activities are carried on by the patients in occupational therapy.

and people elsewhere who are engaged in radiation studies, these studies serve as an admirable stimulus to develop laboratory investigations which may be of use in throwing additional light on known and expected effects of radiation in man.

This year also saw the production of a moving picture which in part during its 25-minute showing gives information on the early reactor experiments, the planning, designing, and construction stages of the medical research reactor, and the development of procedures for its use. This picture, to be used at the Second Geneva Conference on the Peaceful Uses of Atomic Energy, bids fair to have a much more extensive use in this Department as a demonstration of some of its interests.

Another special project was that of designing and having built a model of the medical research reactor which could be used to explain to staff members and visitors various details of the medical reactor. It is planned to locate this model in the lobby of the new building.

#### **HOSPITAL**

The increase in use of hospital facilities for research studies continued during the past year. Patient days totaled 8759, compared with 7327 for the previous year. During the year 93 persons were admitted to the hospital; of these 7 were employees and 86 were research patients. The ambulatory service for patients who could be followed up satisfactorily in this manner in order to reduce their terms of actual hospitalization and whose diagnostic and therapeutic tests could be completed without hospitalization accounted for 436 out-patient visits compared with 376 during the previous year. The total number of persons seen on the research service was 123, of whom 97 were new patients. Radioactive isotopes of 14 different elements were used in the diagnosis and therapy of 74 patients during the year. The major disease states studied and treated in the hospital were glioblastoma multiforme, leukemia, polycythemia vera, rheumatoid arthritis, hypertension, visceral cancer, paralysis agitans, diabetes mellitus, multiple myeloma, and disorders of the thyroid. No services are maintained for specific disease states, since the clinical investigations are basic; each patient, following referral by his own physician, is individually selected both for disease and stage of disease, and is admitted because study or treatment of the disorder at that time

is thought to be advantageous to both patient and physician. In general, the medical profession has been most cooperative in referral of patients to the hospital and in the necessary follow-up studies.

An Emergency Plans Committee has been appointed to recommend and evaluate procedures for maintaining the highest standards of patient safety and welfare in the new Medical Research Center.

#### **INDUSTRIAL MEDICINE**

During the fiscal year visits to the Industrial Medicine service increased in greater proportion than in many recent years. Visits totaled 13,133, compared with 10,692 in the previous fiscal year. Routine annual examinations are being scheduled at greater than 12-month intervals because of the time required for daily volume of loss-of-time prevention visits and the priority scheduling of examinations of employees engaged in radiation areas or in hazardous activities. The number of X-ray examinations increased from 1780 to 2005. All roentgenological examinations are carefully controlled so that a minimum exposure is given. The X-ray equipment obtained for installation in the new Medical Research Center will include an image intensifier with an 8-in. scanning area enabling further reduction of exposure during fluoroscopic examinations. In general, the state of physical fitness of our employees continues to be excellent in relation to the tasks to which they are assigned.

#### **GENERAL ACTIVITIES OF THE MEDICAL DEPARTMENT**

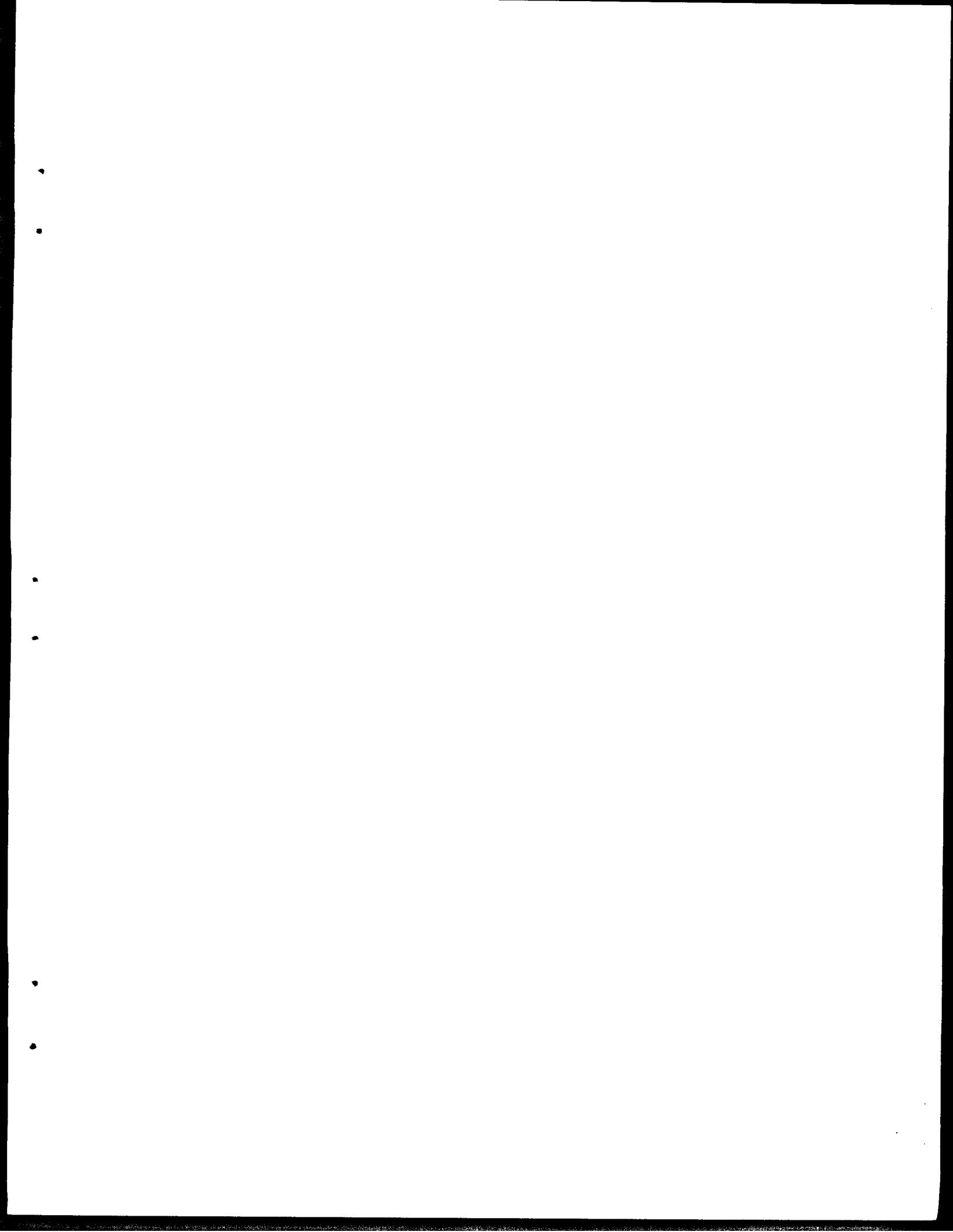
During the fiscal year, a total of 41 staff scientists carried on the work of the Department with an average of 100 technical assistants and 15 administrative and clerical assistants. The program includes work with 70 research collaborators coming from 40 institutions, 12 students from 12 institutions, and 13 visiting scientists from 11 institutions. This year saw our most extensive use of research collaborators; with their help it is possible to amplify and intensify various aspects of the Department's activities.

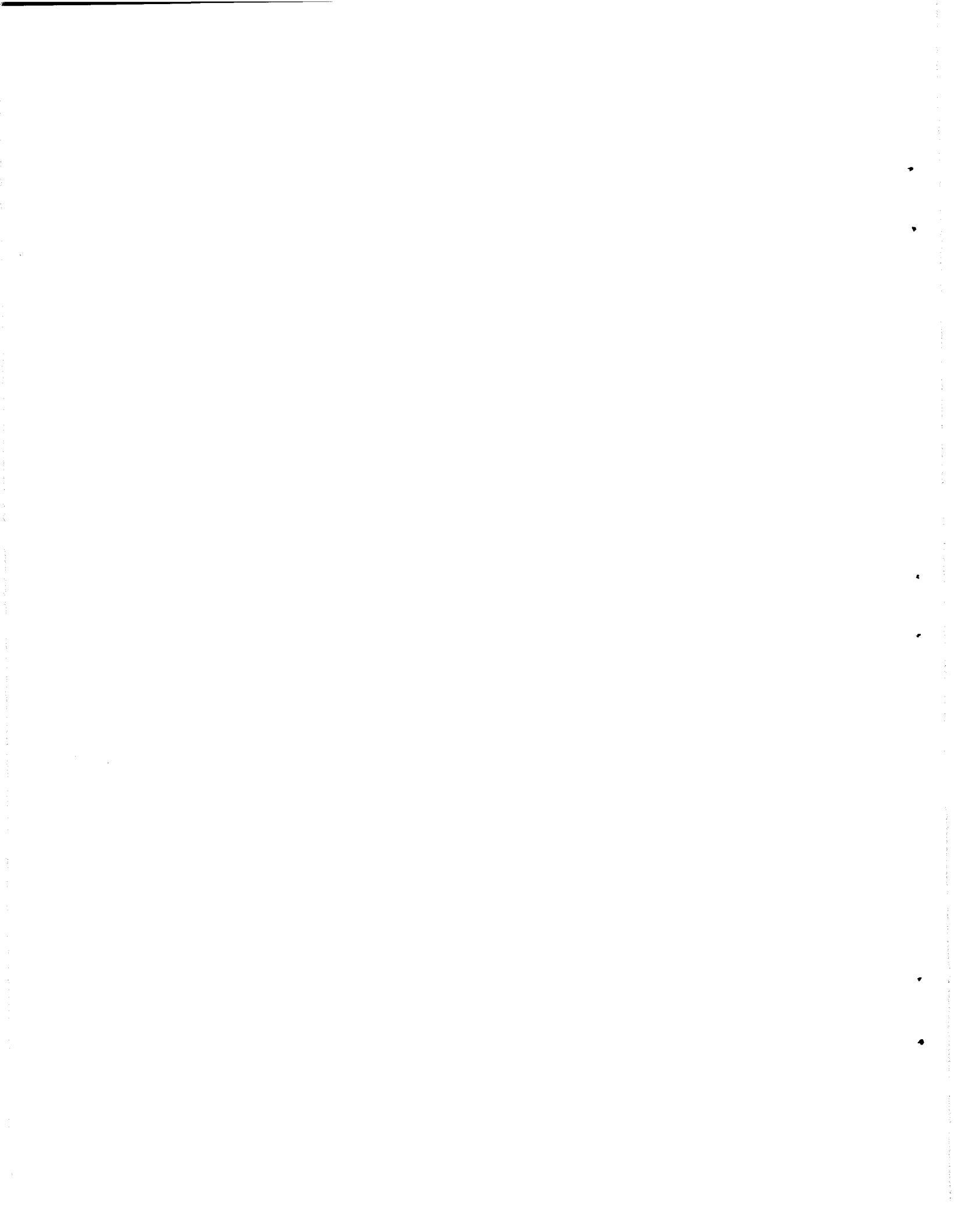
Annually the number of lectures given by the staff increases. During the year 36 different members of the Department gave a total of 159 lectures

or informative talks to groups or societies in 89 different institutions, including institutions in eight foreign countries. These talks ranged from highly technical lectures to broadly informative talks. A very significant educational effort is represented by this activity. In addition, 152 professional visitors from 21 countries participated in personal conferences with members of the Department. This total does not include visitors in groups larger than four members, and the great majority were visiting as single individuals. A great deal of time and effort was consequently needed to meet this demand, which has been estimated as requiring an amount of time at least equal to one man-year. In addition to the individual visitors, five requests were received from scientific groups who wished to visit the Laboratory for a day and to have a program prepared for them. Such a program usually consists of tours of the facilities and three or four lectures with discussions on current research activities. Occasionally the request is for

an educational session, in which instance general orientation talks are given. A total of 320 individuals attended these meetings. In addition, there were three groups of nurses who came here for demonstrations and talks on nursing procedures in relation to patient radioisotope administration.

A special educational activity developed during the last year has been concerned with the needs of the chairmen of departments of the various disciplines in the medical schools with the development of the field of nuclear medicine. As a part of this Department's cooperative efforts with the U.S. Atomic Energy Commission, the latter's Division of Biology and Medicine cosponsored a meeting on February 6 and 7, 1958. It was the second conclave in the series on nuclear energy in medicine entitled, *The Responsibility of the Chairman of the Department of Pathology for Training, Research, and Hospital Practice in the Field of Nuclear Energy*. Chairmen from 73 of the 91 American and Canadian medical schools attended.





## Technical Operations and Services

A number of organizational units in the Laboratory provide the technical services and facilities essential to the research programs. Descriptions of their operations follow under appropriate headings.

### REACTOR OPERATIONS

The reactor was operated on a continuous basis most of the year. The enrichment of the reactor, commenced during 1957, was completed during April 1958. The final phase of the reloading with enriched fuel required a shutdown period of 24 days.

The unscheduled downtime of the reactor during fiscal 1958 totaled 75.2 hr and was the result of 12 accidental or emergency shutdowns. Electrical power failures accounted for 8 of these shutdowns and for 55.6 hr of the downtime. The reactor was in operation at full power for 76.3% of the total time during this period.

### Graphite Annealing

The reactor was annealed twice during the fiscal year, on September 20, 1957, and on May 31, 1958. Because of the questions raised following the Windscale incident in October 1957, the BNL reactor annealing procedures were studied extensively, and a revised procedure was used in the annealing of May 31, 1958. This procedure resulted in a satisfactory operation with respect to safety and release of stored energy, but was less successful than previous anneals with respect to graphite growth recovery. The main difficulty with the revised procedure was connected with the amount of cooling used during annealing. Tests are being made to determine the minimum safe flow of air required during annealing to transport and mix volatile fission products so that they may be promptly detected if inadvertently released. Further revisions in these procedures are to be studied.

### Operation With Enriched Fuel

The replacement of normal fuel with enriched fuel was begun in January 1957, when 35 channels

of enriched fuel were loaded into the central core region. In October 1957 the enriched loading was gradually increased to 120 channels. Between October 1957 and March 1958 the enriched loading was gradually increased to 157 channels. The prolonged shutdown of the reactor from March 20 through April 13, 1958, was for the purpose of removing all the remaining natural uranium fuel from the reactor and adding enriched fuel to provide the required excess reactivity for operating at a central flux of  $2$  to  $3 \times 10^{13}$  neutrons/cm<sup>2</sup>-sec.

When reloading was completed, 240 channels contained fuel, which, on an average, had experienced very little burn-up. Since it was planned to operate the fuel to 40% burn-up, the size of the reactor was small compared to its calculated equilibrium size of 575 channels. Although the central channels have been subsequently operated at essentially the full design power density, the power

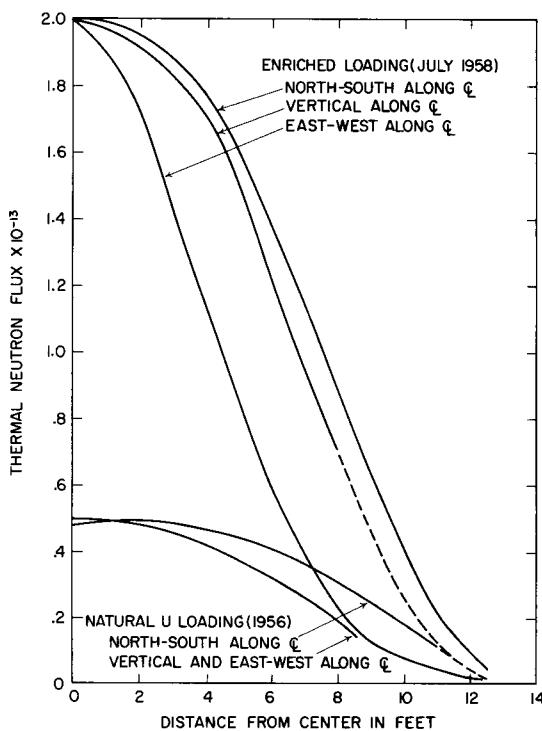
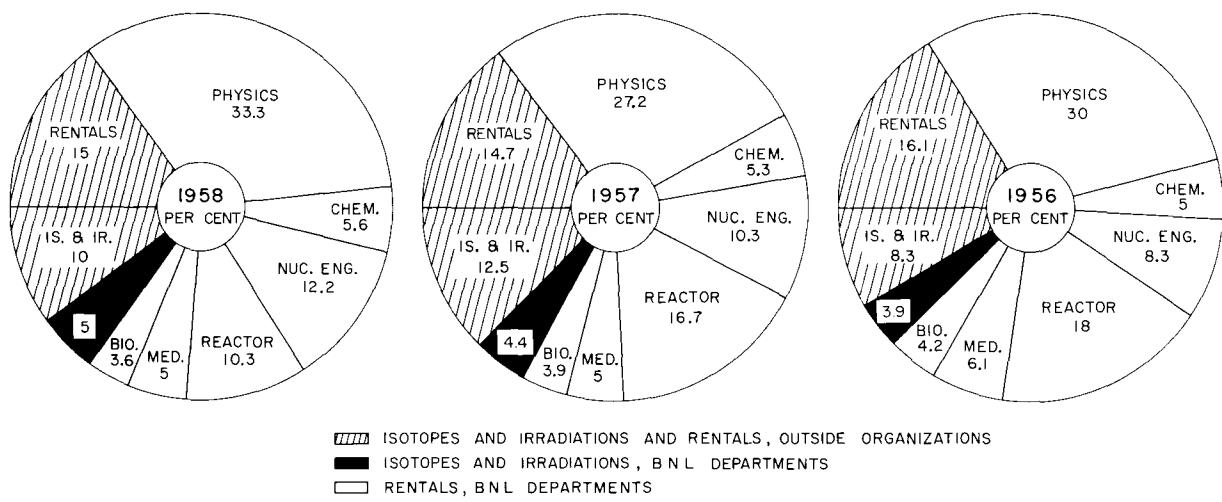


Figure 1. Thermal neutron flux distribution through reactor.

Table 1  
Summary of Reactor Use Charges



	Fiscal Year		
	1958	1957	1956
<b>RENTALS</b>			
Outside Organizations	\$ 197,562	\$ 202,202	\$ 191,125
BNL Departments			
Physics	\$439,898	\$377,788	\$360,396
Chemistry	74,280	74,238	64,245
Nuclear Engineering	159,420	140,698	98,665
Reactor	133,874	229,842	222,638
Medical	66,000	70,760	66,816
Biology	46,754	920,226	44,234
Total	\$1,117,788	\$1,147,650	\$1,048,119
<b>ISOTOPES AND IRRADIATIONS</b>			
Outside Organizations	\$ 130,931	\$ 172,801	\$ 100,523
BNL Departments			
Physics	\$12,121	\$18,759	\$15,991
Chemistry	9,114	6,910	5,258
Nuclear Engineering	37,620	28,264	26,254
Reactor	1,250	1,646	1,936
Medical	4,973	4,903	5,221
Biology	3,474	1,122	698
Other	398	475	464
Total	\$ 199,891	\$ 234,880	\$ 156,345
Total use charges	\$1,317,679	\$1,382,530	\$1,204,464

output of the reactor was initially low, 10 Mw. As burn-up has progressed, additional fuel has been added to maintain reactivity, and the power has increased. As of July 1, 1958, 292 channels were loaded, and the power output was 13 Mw. The

central flux has increased slightly as the reactor became larger. In Figure 1 the thermal flux as of July 1958 and the thermal flux of the natural fuel loading are plotted to show the thermal flux enhancement.

Table 2  
Three-Year Summary of Irradiation Services for Outside Users

	Fiscal year					
	1958		1957		1956	
	Number	Volume, \$	Number	Volume, \$	Number	Volume, \$
Reactor irradiations	1,153	66,683	1,175	60,822	1,177	61,694
Special isotopes	200	32,021	185	15,951	85	7,265
Research subsidy	—	(19,649)	—	(10,812)	—	(4,788)
Co <sup>60</sup> sources	8	29,753	13	93,282	7	33,560
Cyclotron irradiations	14	2,836	18	3,047	16	3,223
Gamma irradiations	—	29,553	—	27,000	—	56,000
Total	1,375	160,846	1,391	200,102	1,285	161,742

### Utilization of the Reactor

Insofar as is practical, the reactor's experimental facilities have been fully utilized for several years. Some additional usefulness has been realized from time to time by conducting experiments that require almost no pile room floor space in reactor holes that otherwise would not be used because of limited access. A recent illustration was the utilization of a fuel channel for a HNO<sub>3</sub> loop, whose out-of-pile portion was located in another building, the Hot Laboratory. The relative use of the reactor by the BNL research departments and outside research organizations for the past three years is indicated in Table 1.

Given below is a list of the outside organizations renting experimental holes during the past year, together with a brief statement characterizing their work:

American Bosch Arma Corporation (government contract)  
Radiation damage on electronic circuits  
American Nuclear Science Corporation (government contract)  
Radiation damage on diodes  
American Machine & Foundry Company (government contract)  
Radiation damage on capacitors  
Bell Telephone Laboratories, Inc.  
Neutron spectrometer (utilization agreement)  
Radiation damage on electronic components (government contract)  
Radiation damage to ferrite cores (government contract)  
Esso Research and Engineering Company (private funds)  
Irradiation of hydrocarbons under controlled conditions of temperature and pressure  
Ford Instrument Company (government contract)  
Fuel element study for gas cooled reactor

General Dynamics Corporation (utilization agreement)  
Neutron spectrometer  
General Electric Company  
Neutron spectrometer (utilization agreement)  
Radiation damage tests on electronic components (government contract)  
Naval Ordnance Laboratory  
Radiation damage on magnetic materials  
Naval Research Laboratory  
Radiation damage on steel alloy test specimens  
Pennsylvania State University (utilization agreement)  
Neutron spectrometer  
Rensselaer Polytechnic Institute (government contract)  
Nitrogen fixation experiment  
Sperry Gyroscope Company (government contract)  
Viscosity changes in certain fluids due to radiation damage  
Radiation damage test on electronic components  
Westinghouse Electric Corporation (government contract)  
Fission gas diffusion experiment  
Radiation damage to steel test specimens  
Investigation of the phase change in uranium based alloys due to radiation damage  
Operated a neutron spectrometer under a usage agreement with the Laboratory

### Isotope Production and Irradiation Services

Table 2 presents a 3-year summary of the volume and income involved in irradiation services performed for outside organizations.

The number of reactor irradiations and income therefrom have remained relatively constant but should increase as higher fluxes become available.

The production of special-process radioisotopes by the Hot Laboratory (see Table 3) has shown a pronounced increase, and the income therefrom has doubled each year. Of the radioisotopes at present involved, Mg<sup>28</sup> accounts for ≈63% of the

Table 3  
Shipments of Special Isotopes

Isotope	Fiscal year			
	1958	1957	Number of shipments	Activity, mC
I <sup>132</sup>	45	2020	73	2336
I <sup>133</sup>	30	1030	38	820
Mg <sup>28</sup>	130	19.8	73	8.82
F <sup>18</sup>	6	80	9	50.5
Y <sup>90</sup>	2	1400	1	100

volume and 78% of the dollar income. I<sup>132</sup> ranks second with  $\approx 22\%$  of the volume and 17% of the dollar income. The doubling of the total dollar income over last year without a significant increase in total shipments was due to a large increase in the demand for high priced Mg<sup>28</sup>.

Under the AEC Research Support Program, an 80% discount on the irradiation service charge is available in the case of certain medical and agricultural research. The amount of research subsidy borne by Brookhaven for such applications has roughly doubled each year, as shown in Table 2.

The amount of Co<sup>60</sup> shipped during fiscal 1958 was  $\approx 12,000$  curies or 20% less than last year. The disproportionate drop in income was due to a decrease in the price for Co<sup>60</sup> from \$5 to \$2 per curie. In addition,  $\approx 1800$  curies were issued under AEC agreements and as make-up activity at no charge.

The use of the BNL 60-in. cyclotron for irradiations for outside users has always been of a minor nature inasmuch as process cyclotron-produced radioisotopes are available commercially. The few cyclotron irradiations performed by BNL are connected with cooperative programs with universities or other federal agencies.

The AEC is encouraging the use of commercial gamma irradiation facilities whenever possible. To some extent this has curtailed BNL gamma service irradiations for outside organizations. However, the income continues to remain about the same.

#### Procurement of Special Materials

The procurement for the scientific departments of all radioactive and stable isotopes as well as special materials and services controlled by the

AEC is the responsibility of the Isotopes and Special Materials Group. In this connection, 167 purchase orders were placed for radioisotopes, 26 for stable isotopes, and 43 for special materials. This service was utilized chiefly by the Medical Department (over 30%), the Chemistry and Biology Departments (25% each), and the Physics Department (15%).

A new irradiation program was initiated at the National Reactor Testing Station, Idaho Falls, Idaho, for the Nuclear Engineering Department. This program involves the production of a megacurie of Co<sup>60</sup> during the next year or so in the new Engineering Test Reactor. Five other irradiation programs in the Materials Testing Reactor were completed during the year, which leaves three programs still under way.

The Isotopes and Special Materials Group has also maintained an inventory of the enriched stable isotopes at Brookhaven procured by purchase and loan. During the year an inventory was also made of the radium sources on hand, and these sources were leak-tested.

#### Source and Special Nuclear Materials Accountability

Table 4 presents a 3-year summary of the amounts of source and special nuclear materials on hand at BNL at the end of each fiscal year.

The refueling of the research reactor with enriched uranium increased the inventory of U<sup>235</sup> of over 75% enrichment, as did the procurement of enriched fuel for the Medical Research Reactor and for the critical experiments being performed by the Nuclear Engineering Department.

Table 4  
Source and Special Nuclear Materials on Hand  
at End of Year (in kilograms)

	Fiscal year		
	1958	1957	1956
Natural U	9,810	54,127	63,642
Depleted U	45,955	11,745	11,263
U <sup>235</sup> over 75%	163	51	13
U <sup>235</sup> under 75%	4,776	3,679	3,853
U <sup>233</sup>	0.1	0.1	0.1
Pu <sup>239</sup>	21	7	7
Th	194	169	169
Heavy water	176	178	61

The large amount of natural uranium present last year as reactor fuel was shifted to the category of depleted uranium when it was discharged from the reactor.

As an AEC contractor, the Laboratory submits monthly inventories of each of the listed materials. Both the materials and records are subject to an annual audit by the AEC; the last audit was conducted in May 1958. The present manual of accountability procedures is being revised to incorporate changes necessitated by the problems of criticality control resulting from the increased amount of fissionable material at the Laboratory.

Based on dollar values published by the AEC, the current inventory of the materials listed in Table 4 represents a value of  $\approx \$7,700,000$ .

### Medical Research Reactor

The housing and shielding of the Medical Research Reactor are currently being constructed as a part of the new Medical Research Center by the Malan Construction Company, the general contractor for the entire Center, under AEC supervision.

The components of the reactor proper have been procured directly by the Laboratory and with minor exceptions the fuel, reactor vessel, control rods, instrumentation, and special materials were on hand as of January 1, 1958. Installation of many of these items awaits completion of the shielding and housing, and two to three months more will be needed before the reactor becomes operational. One Laboratory staff member has been engaged full time in inspecting and otherwise assisting the AEC during the construction of the Medical Research Reactor.

### HOT LABORATORY OPERATIONS

Besides the production and processing of special isotopes and the gamma irradiations described above and listed in Tables 2 and 3, the Hot Laboratory staff provides many other services, some developmental in character, which are briefly described below.

#### Radiochemical Analysis

Chemical analyses of radioactive materials were performed on a service basis, and in addition the following procedures were developed or modified during the year.

- 1) Analysis of trace amounts of Th in the presence of large amounts of Bi by using a liquid-liquid extraction with tri-isoctylamine to complex and remove the Bi.
- 2) Determination of small amounts of Al in steel containing Ti, by the chloroform extraction of Fe and Ti chelates formed with 8-hydroxyquinaldine.
- 3) Coulometric determination of U and Mo in the presence of each other by using electrolysis at controlled cathode potentials.
- 4) Polarographic determination of U in Bi without prior separation.
- 5) Determination of Ta in Bi by activation analysis.
- 6) Spectrographic determination of Ta by using Nb as both an internal standard and a carrier for the Ta.
- 7) Chronopotentiometric analysis of the  $MgCl_2$ - $NaCl$ - $KCl$  ternary eutectic.
- 8) Determination of  $<10$  ppm U in ternary  $MgCl_2$ - $NaCl$ - $KCl$  eutectic, by extraction of U into tris(ethylhexyl)phosphine oxide followed by direct fluorimetric analysis.
- 9) Analysis of Xe adsorbed on or beneath the surface of graphite, by complete combustion of the C and subsequent adsorption of the Xe on cold charcoal.
- 10) Spectrographic and colorimetric procedures for Cu.
- 11) Determination of  $Pa^{233}$  by scanning a solid sample with a gamma spectrometer.
- 12) Assay of  $BiCl_3$  by precipitation of the sub-carbonate.
- 13) Colorimetric determination of Mg.
- 14) Colorimetric determination of iodide.

Methods for the routine analyses of a variety of elements and compounds have also been standardized.

#### Other Activities

A total of 1759 routine radiographs were made during the year, most of them in connection with the Liquid Metal Fuel Reactor investigations.

The new metallurgical hot cell was designed with the aid of a full-size mock-up of  $\frac{1}{3}$  of the cell, and construction was started. A smaller-scale shielded bench and temporary equipment were set up for use in metallurgical testing of hot samples.

The activities of the Hot Laboratory also included:

- 1) Mock-up of four motions of the BNL Model 4 rectilinear manipulator, which were found to work satisfactorily.
- 2) Design and construction of a new type of lead shield for the 100-mC  $I^{132}$  generator.
- 3) Reconditioning of manipulators, periscopes, etc. damaged by the explosion on May 15, 1957.
- 4) Improvements in the design of the electrical system of the remotely operated medical infusion pump.
- 5) Construction of a new arc chamber sample holder for the emission spectrograph.
- 6) Development and construction of equipment for the new  $Te^{132}$  separation process.
- 7) Repair and reloading of a 150-curie  $Co^{60}$  irradiator.

#### Waste Processing

At the end of the second quarter the "D" waste inventory stood at 223,000 gal and was increasing at the alarming rate of  $\approx 30,000$  gal/month. The Waste Concentration Plant was put on 2-shift operation for several months in order to reduce this inventory. Table 5 indicates the magnitude of the liquid waste operations for the past two years.

#### HEALTH PHYSICS

The responsibilities of the Health Physics Division were increased somewhat by the addition in July 1957 of the general (nonradiation) safety program, previously the responsibility of the Personnel Division. As in the past, most of the major problems of exposure and contamination control were encountered at the nuclear reactor and in waste disposal areas. These problems were especially difficult this year because of the reloading of the reactor with new fuel elements. The Cosmotron also presented acute problems in exposure control prior to its shutdown in November.

Reloading of the pile with enriched fuel has had several implications in regard to radiation safety. Because of the higher flux levels, objects removed from the pile will be more radioactive and therefore more difficult to handle. The radioactive wastes resulting from operation and use of the pile will be considerably "hotter," a fact which has already strongly affected the activities of the waste disposal group. Five on-site area monitoring stations have been reactivated to aid in evaluating

Table 5

Liquid Waste Operations				
"F" Waste Discharged to the Sewer				
Period	Volume, gal	Average activity level, C/ml	Total activity, mC	
Fiscal 1958	4,667,244	$1.95 \times 10^{-11}$	343.6	
Fiscal 1957	4,978,335	$1.77 \times 10^{-11}$	352.6	
Waste Concentration				
Feed, gal			557,969	
Average volume reduction			118:1	
Drummed concentrate, gal			4,707	
Waste Inventory in Gallons				
	Fiscal 1958		Fiscal 1957	
	"D" waste	"A" waste	"D" waste	"A" waste
Volume on hand at beginning of fiscal year	158,545	3592	99,310	2927
Volume received during year	492,280	168	463,795	665
Volume to evaporator	556,805	—	404,560	—
Volume on hand at end of fiscal year	94,020	3760	158,545	3592

the increased  $A^{41}$  output from the reactor stack. Levels both on and off the site appear to be well below the prescribed limits, although at times peak levels that interfere with the counters in some of the laboratories are encountered.

As usual, a group of AEC Fellows in Radiological Physics is at the Laboratory for field training in health physics this summer. Approximately 50 individuals from 12 foreign countries have received training in health physics this year for periods ranging from a few hours to a full year. About twice as many domestic visitors and trainees have been similarly accommodated.

Routine measurements of the rate of deposition of radioactivity in rainfall and settled dust were continued by means of the pot collection method. The average rate for the year was 6180 dis/min/ $m^2/day$ . The highest value for a single day

(148,000 dis/min/m<sup>2</sup>/day) was encountered in September.

### Personnel Monitoring

The use of personnel monitoring equipment, shown in Table 6, has declined somewhat, presumably because of the protracted shutdown of the Cosmotron. Exposure totals and averages for the 1483 individuals monitored are shown in Table 7, and Table 8 shows the distribution of individual exposures by magnitude, in terms of yearly values and of values integrated over the total employment period. In 1957 as compared to 1956, the average exposure per individual dropped 15%, and there were only 12 exposures over 5 r (equivalent) as compared with 18. None of the integrated exposures is in excess of the value corresponding to an average of 5 r per year.

### General Safety

The general (nonradiation) safety program was transferred to the Health Physics Division in July 1957. L.A. Baker, Jr., a professional safety engi-

neer, was appointed Manager of Safety Services. The staff has been augmented recently by addition of a second safety engineer who will act as safety representative to the various service divisions, make inspections in connection with the construction safety program, and conduct accident investigations.

To implement the field work of the Safety Services Office, the health physics surveyors were assigned collateral duties as safety representatives to the various departments. Training in this work has been conducted at monthly seminars as well as on an informal basis. Department chairmen were asked to delegate their safety responsibilities to department safety coordinators. This arrange-

Table 8

Distribution of Exposures of Individuals Receiving Regular Personnel Monitoring Service

Table 6		
Use of Personnel Monitoring Equipment		
	Fiscal 1958	Fiscal 1957
Pairs of pocket chambers	913	919
Regular film badges	614	734
Visitors' film badges - on-site	82	96
Visitors' film badges - off-site	193	198
Total film badges	889	1029
Neutron films used	578	762
Neutron films read	315	280

Figures are weekly averages for each fiscal year.

Type of exposure	Total for all individuals	Average	
		Average per individual	per individual per week
$\beta$	166 rad	0.112	0.0021
$\gamma$	222 r	0.149	0.0029
$n$	83 rem	0.056	0.0011
E.I.*	388	0.294	0.0056

\*E.I. = Exposure Index ( $\frac{1}{2}$  of  $\beta$  exposure in rads +  $\gamma$  exposure in r + neutron exposure in rem).

Exposure interval*	No. of persons receiving stated exposure		
	Calendar 1956	Calendar 1957	Integrated**
0.0- 0.5	1223	1298	976
0.5- 1.0	151	93	121
1.0- 1.5	48	25	62
1.5- 2.0	30	11	42
2.0- 2.5	12	18	37
2.5- 3.0	12	10	26
3.0- 3.5	9	4	37
3.5- 4.0	5	6	24
4.0- 5.0	7	6	35
5.0- 6.0	8	4	30
6.0- 7.0	2	4	11
7.0- 8.0	4	2	10
8.0-10.0	4	2	20
10.0-12.0	0	0	11
12.0-15.0	0	0	8
15.0-20.0	0	0	9
20.0-25.0	0	0	9
25.0-30.0	0	0	7
30.0-40.0	0	0	5
40.0-50.0	0	0	2
50.0-60.0	0	0	1
Total	1515	1483	1483

\*Tabulation is in terms of values of exposure index as defined in Table 7.

\*\*No. of persons receiving stated exposure during their total BNL employment period.

ment greatly simplifies dealing with the various departments in safety matters.

The accident investigation system has been reorganized. A report of any injury in the first-aid category is provided to the department safety coordinator for appropriate action within the department. More serious injuries are investigated jointly by the supervisor, the department safety coordinator, and the Safety Services Office, and means of preventing recurrence are determined by this group. The circumstances of each incident are analyzed, and the data thus obtained are used as a guide for future program emphasis.

Periodic inspection in each scientific department is now arranged by the department safety coordinator and conducted by department personnel with the assistance of the safety representative. The department safety coordinator verifies the elimination of reported hazards or unsafe procedures, with the assistance of the Safety Services Office in the case of unusual problems.

The accident rate during the past six months has shown substantial improvement, the number of disabling injuries being 3.2 per million man-hours (per A.S.A. Code Z 16.1), about half the rate for the preceding year. This improvement may be due in part to the initial spurt connected with reorganization of the safety program, and vigilance must be continued if a low accident rate is to be maintained.

### Research and Development

Most of the research and development activities in the Health Physics Division are closely related to radiation protection problems. Three new instruments have been developed during the past year. The first is a giant beta-sensitive ionization chamber for use in checking protective garments such as lab coats or coveralls after laundering. The dimensions of the chamber are 5 ft  $\times$  2½ ft  $\times$  4 in., and its front face is ½-in. expanded steel mesh covered with aluminum foil. The value of this type of monitor lies in its ability to respond quantitatively to contamination regardless of its distribution on the garment. Full reading on the most sensitive scale of the associated electrometer corresponds roughly to 1  $\mu$ C of relatively old fission product contamination.

The second instrument developed is an automatic grab sampler and decay plotter for use at the pile. When one of the continuous dust monitors routinely operated in the pile building in-

dicates an abnormal level of airborne activity, a valve opens to an evacuated tank attached to the sampling head, and a sample of the atmosphere is quickly obtained. The activity of this sample is immediately measured and plotted as a function of time by an associated counter and ratemeter. This equipment is very useful in determining the nature and magnitude of unexpected particulate activity.

The third instrument is a new type of neutron spectrometer developed by a visiting German health physicist working in collaboration with a member of the Physics Department. The spectrometer makes use of two scintillation counters having KI and NaI crystals, respectively. The incident neutrons initiate the  $K^{39}(n,p)A^{39}$  reaction in the KI crystal. The pulses caused by the proton group leading to the 1.27-Mev first excited state of A<sup>39</sup> are isolated by means of the coincident NaI pulses caused by the de-excitation gamma-rays from this state. In this way output pulses due to a proton group whose energy distribution is linearly related to that of the incoming neutrons are obtained. The spectrometer has an efficiency of  $\approx 5 \times 10^{-5}$  and a neutron resolution of  $\approx 12\%$  for 4-Mev neutrons.

Two methods for determining the effective relative biological effectiveness (rbe) of the mixed radiation encountered in working areas at the Cosmotron were under investigation prior to its shutdown in November. The first method depends on a quantitative analysis of tracks in a diffusion cloud chamber. The lengths of tracks with four ranges of linear energy transfer (LET) are determined and suitably weighted for the energy released per unit length and the rbe appropriate to each range of LET. By using the table in *NBS Handbook 59* that relates rbe to LET, a value of effective rbe may be obtained. However, this method has been abandoned, since it is tedious and inaccurate, and would require a cloud chamber with a minimum of metal parts to give results of biophysical significance. In the alternative method, a so-called LET chamber lent by H. Rossi of Columbia University is used, which is a spherical tissue-equivalent chamber. By analysis of the pulse height distribution obtained with the LET chamber a curve is derived showing the distribution of absorbed dose with LET. Since the total absorbed dose at the chamber location is independently measured and a rough relationship between LET and rbe is available, the effective

rbe may be computed. Reasonable preliminary values were obtained at two locations, but much work remains to be done when the Cosmotron is again in operation.

Measurements of the radiation levels in a well used by the Physics Department for storing nuclear track films were made with a NaI crystal and counter. It was found that the background level in the well was less than  $\frac{1}{3}$  of the normal surface values; moreover, it was very little affected by surface disturbances such as a tenfold increase in level caused by passage overhead of A<sup>41</sup> from the pile. It was established that a further reduction by a factor of 2 could be effected by a 1-in.-thick steel shield, the thickest that could be accommodated.

#### Waste Disposal and Reclamation

A full one-year accumulation of waste packaged in 55-gal drums was trucked to Floyd Bennett Field early in July 1958 and dumped at sea by a Navy LST. The shipment consisted of 692 drums with an estimated activity content of 1000 to 1200 curies. It is apparent that two shipments a year will be needed in the future and that the practical upper limit of activity per drum has been nearly

reached. The amount of active slurry received from the evaporation plant increased substantially. However, it was found possible to make concrete in the mixer with slurry and pure cement, and thus to package more slurry per barrel and prevent a large increase in the number of barrels required. Use of transit-mix concrete for topping and lining barrels has reduced personnel exposures during these particular operations. Exposures in connection with the hot concreting operation have been greatly reduced by provision of an underground tank for storage of the hot slurry.

Data relating to the liquid waste system are presented in Table 9. A modest increase in the activity released is indicated. However, the total released to the filter beds for the year (0.32 curies) is only 20% of the official 1.5-curiel limit. The year's heavy rainfall (56 in.) diluted the waste effluent crossing the site boundary to an unprecedented degree.

About half of the time of the five members of the health physics waste group was devoted to reclamation activities. Improvement of the wet decontamination facility for cleaning fuel storage tubes from the pile reduced the time required per tube to  $\frac{1}{3}$  or less of the previous value, and personnel exposures were far lower than previously. A peak load of work was experienced at this facility in connection with the pile reloading.

Other items reclaimed from the pile included orifice plugs, heat exchanger units, shipping containers, and magnets used in the canal. A procedure was worked out whereby filter cartridges used in respirators can be sterilized and reused. Since these units cost more than a dollar each and 300 or more are used per month, a considerable annual saving will result from the use of this procedure.

#### METEOROLOGY

In addition to performing its customary service functions, including various climatological studies, analysis of diffusion problems, and a limited number of routine forecasts, the Meteorology Group completed an automatic system of predicting the diffusion of the cooling air from the reactor in the spring of 1958. This system actually is a synthesis of a number of developments by Health Physics and Meteorology personnel. Reactor operations personnel now have available in the control room continuous records of the radioactivity present in the cooling air at various points in the exhaust

Table 9

#### Summary of Liquid Waste Data

	Fiscal 1958	Fiscal 1957
Input to filter beds, gal/day	467,500	427,500
Output from filter beds, gal/day	368,800	332,000
Net loss in filter beds, %	21.1	21.4
Stream above discharge point, gal/day	934,400	315,000
Stream at site boundary, gal/day	1,460,000	778,500
Rainfall, in./mo	4.7	2.9
Activity concentration at input to filter beds, C/cc	$4.5 \times 10^{-13}$	$3.9 \times 10^{-13}$
Activity concentration at output from filter beds, C/cc	$1.8 \times 10^{-13}$	$1.6 \times 10^{-13}$
Reduction in activity concentration, %	60	58
Activity at input to filter beds, mC/mo	26.7	17.5
Activity at output from filter beds, mC/mo	8.2	6.0
Percent of permissible	19.2	12.5

Figures are averages for each fiscal year.

system, and of wind direction, speed, and gustiness. From these parameters the operator can determine where the cooling air is reaching the ground and approximately what concentrations of radioactivity may be present, without assistance from Health Physics or Meteorology personnel.

This system does not include estimates of deposition of particulates or of their rainout on the ground. However, these variables will be included in the reasonably near future, on completion of a machine program for their calculation, now in the "debugging" stage. The system will also be extended to the Medical Research Reactor when it goes into operation.

#### APPLIED MATHEMATICS - COMPUTATION SERVICES

During the year a major change in emphasis took place in the Division, reflecting an attempt to bring the most modern and advanced computing equipment and techniques within effective reach of the Laboratory's staff. Since July 1957, when an IBM 704 computer became available at the AEC Computing Facility at New York University, several lectures on its use have been given at Brookhaven by Facility personnel, and members of this Division have provided extensive assistance to individual users. Table 10 shows the distribution of use of this machine by departments since operation started. In order to facilitate the present use of this machine and to provide for an expected use of 500 hr during the coming year, several members of this Division have been stationed at New York University.

Table 10

#### Distribution of IBM 704 Time by BNL Departments

Department	Fiscal 1958	
	Usage, hr	Usage, %
Accelerator Development	18	22
Applied Mathematics	6	7
Chemistry	19	23
Cosmotron	5	6
Nuclear Engineering	12	15
Physics	21	26
Reactor	1	1
Total	82	100

Table 11

#### Distribution of Univac Time by BNL Departments

Department	Fiscal 1958		Fiscal 1957	
	Usage, hr	Usage, %	Usage, hr	Usage, %
Accelerator Development	48	15.0	399	48
Applied Mathematics	16	5.0	0	0
Chemistry	26	8.0	25	3
Nuclear Engineering	223	70.0	383	46
Physics	5	1.7	8	1
Reactor	1	0.3	17	2
Total	319	100.0	832	100

Table 12

#### Distribution of 409 Facility Time by BNL Departments

Department	Fiscal 1958		Fiscal 1957	
	Usage, hr	Usage, %	Usage, hr	Usage, %
Accelerator Development	10	1.0	0	0
Biology	7	0.7	63	5
Chemistry	0	0	25	2
Cosmotron	288	30.7	188	15
Health Physics	38	4.0	12	1
Medical	33	3.5	163	13
Nuclear Engineering	413	44.3	351	28
Physics	148	15.8	301	24
Reactor	0	0	150	12
Total	937	100.0	1,253	100

Table 13

#### Assignment of Mechanical Engineering Division Personnel, July 1, 1958

	Designers and Engineers	Secretaries and Draftsmen	and Clerical
Accelerator Development			
Department	12	15½	2
Central Design Group	0	8	1
Cosmotron Department	8	4	0
Nuclear Engineering Department	3	10	1
Physics Department	1	5	0
Reactor Division	0	2	0
Total	24	44½	4

Principally as a result of the availability of the IBM 704 computer, the use of the Univac machine at New York University has decreased to 319 hr from a total of 832 hr during the previous year. The 409 punch card machine on site showed a similar, though less severe, reduction in use, from 1253 hr to 937 hr, and this trend is expected to continue. The distribution of the use of these machines for the past two years is shown in Tables 11 and 12.

### MECHANICAL ENGINEERING

The Mechanical Engineering Division has continued its support of research and engineering projects throughout the Laboratory. Members of the Division have been assigned to several departments (see Table 13) and have participated in the conception, design, and development of equipment as well as in the writing of specifications, analysis of bids for procurement, and inspection and supervision at the plants of fabricators. Division personnel have also assisted in the assembly, testing, and placing in operation of equipment at the Laboratory.

The members of the Division assigned to the Accelerator Development Department have been engaged in the production of equipment for the Alternating Gradient Synchrotron. The design of components has continued, with emphasis on the linear accelerator injector.

Division personnel assigned to the Nuclear Engineering Department have carried out work for that department, the Reactor Division, and the Chemistry Department. Major activities have centered on pumped loops, chemical process vessels or pilot plants, and mechanical components for use in the LMFR program. In addition, the mechanical aspects of the LMFRE research, development, and design work being performed by the Babcock & Wilcox Company for the AEC have been reviewed. The Division has had a major role in establishing operating procedures for the 4-in. utility test loop.

Work was started for the Reactor Division on the modifications necessary to reduce the operating power for cooling the research reactor, and equipment was designed and procured for use by the Chemistry Department in a neutrino detection experiment.

Division personnel assigned to the Cosmotron Department have designed, set into production,

and started to assemble a completely new coil for the Cosmotron. New radiation shielding has been designed and contracts let for its manufacture. Specifications have been written for mechanical equipment in the new target building, and liaison has been maintained with the Architect Engineer during construction. Work has been carried on to aid the Cosmotron Department in increasing the beam intensity and the beam utilization. Members of the Division have supervised the operation of the hydrogen liquifier facility.

Division personnel at the Physics Department devoted their efforts to the construction of the 20-in. hydrogen bubble chamber.

The Central Design Group has assisted personnel assigned to several departments, and has performed design work directly for some other departments. Equipment and components have been designed for use with the research reactor.

Division personnel have been assisting the Reactor Division in the installation of the Medical Research Reactor, the design of equipment for test and research projects on the research reactor, and the establishment of concepts for the proposed high flux research reactor.

### MACHINE SHOPS

The experience of the year past has made apparent a significant change in the demands being placed on the Machine Shops. The major characteristics of this change are twofold: a very substantial increase in the physical size of the larger projects being constructed, and a marked increase in both the quantity of welding work called for and the quality of welding demanded. The general increase in work output required of the Shops has continued in fiscal 1958, with productive hours showing an increase of 7.5% over fiscal 1957. Indications are that this trend will continue in fiscal 1959 and that the productive hours may reach an estimated high of 135,000 hr or almost a 60% increase over the 1955 level.

These changes have called for a fluid and dynamic machine tool equipment policy, as well as continual reappraisal of plant layout in terms of improved efficiency and room required for the indicated expansion of facilities. The second major plant layout of the Shops facilities was carried out during the year, and plans were drawn for additional changes in the coming year. Fiscal 1959 will see the third major plant layout, which will

complete the basic physical reorganization of Machine Shops facilities.

In connection with improvements made in physical layout, steps were taken to alleviate the materials handling problem. Jib cranes were designed, built, and installed at the most critical locations. This has greatly reduced the safety hazards involved and the time required for handling operations.

Although in fiscal 1957 the fabrication requirements of the Laboratory seemed to have stabilized sufficiently to make possible a comprehensive machine tool procurement and retirement plan, it is now apparent that requirements are still in a state of flux, and may remain so for some time. In keeping with the changing demands, a better balance of machine tools and equipment was sought. The acquisition of a planer has filled a major gap in facilities. A radial drill with an arm length of 60 in. was acquired for the handling of larger work. Still much needed are larger size milling machines and additional lathes.

The organizational structure remains the same as in fiscal 1957. It provides good control of overall operations, yet gives first-line supervisors complete authority to carry out their responsibilities.

The productive work output of the Machine Shops consisted of work formally requisitioned

( $\approx 93\%$ ) and short-order work ( $\approx 7\%$ ). Slightly more than 75% of the workload was handled by the machining group, and the remainder by the sheet metal and welding group.

The contributions of the several departments to the workload are indicated in Table 14. The distribution of load between major and minor users remains approximately the same as last year, although the relative contributions of individual departments show considerable change.

The Machine Shops have provided the services of men to the Chemistry, Physics, Accelerator Development, and Nuclear Engineering Departments on a loan basis. Such services have accounted for  $< 5\%$  of the productive man power during fiscal 1958, but are expected to increase in fiscal 1959 to as high as 20% as a result of increased demand by the Accelerator Development and Cosmotron Departments.

## TECHNICAL INFORMATION

As a result of the recent declassification by the AEC of further areas of research, the shift from classified to nonclassified work at Brookhaven has continued. Now that only a few minor phases of the program must be conducted on a classified basis, the Laboratory's technical information functions have undergone a major change. The steady decrease in the volume of holdings that must be safeguarded in the Classified Library has entailed a proportional increase in the activities of the Research Library, with concomitant shifts in staff. A further index of the trend toward unclassified research in all areas is the fact that not a single classified report has been published during the current year. In fiscal 1957 two reports were published on a classified basis, but have since been declassified. In the preceding year 14 such reports were published, and for fiscal 1955 the figure was 22.

In the Research Library, figures for book acquisitions rose slightly above last year's, and a substantial number of new journal subscriptions, particularly in the field of nuclear engineering, were added to the collection, which now totals over 900 titles. Several sets of back issues of journals in the fields of biology and medicine were also acquired.

In addition to its normal functions, which have increased by 25% during the past five years, the Library's circulation section has been assigned the

Table 14

### Departmental Contributions to Shops Workload

	Fiscal 1958 % ILR man-hours	Fiscal 1957 % ILR man-hours
<b>Major users</b>		
Nuclear Engineering	40.8	36.2
Physics	19.3	22.2
Reactor	13.0	15.5
Cosmotron	6.0	9.5
Accelerator Development	5.2	2.9
Percent of workload	84.3	86.3
<b>Minor users</b>		
Medical	4.6	4.3
Chemistry	4.0	2.1
Instrumentation and		
Health Physics	2.3	0.8
Biology	1.5	2.6
Miscellaneous	3.3	3.9
Percent of workload	15.7	13.7

responsibility of issuing and receiving laboratory notebooks and conducting an inventory of these workbooks. This increased workload has resulted in the addition of a temporary employee for the summer months.

In the reference section the effects of the declassification program continued to be evident in the increased volume of declassified and unclassified reports and accompanying catalog cards. The *Weekly Selected Reading List*, compiled primarily for the Laboratory's scientific staff but also used extensively at other nuclear energy installations both here and overseas, has now entered its eleventh year. Each week over 1600 copies of this 8 to 10-page survey of current atomic energy literature are distributed. A number of bibliographies were prepared for staff members during the past year, including the extensive *Bibliography on the Effects of Ionizing Radiations on Plants, 1896-1955* (BNL-504) compiled in cooperation with members of the Biology Department's staff. As part of a nationwide program to disseminate technical information emanating from countries in the Soviet orbit, the Library continues to review Soviet scientific monographs and laboratory reports, and to recommend to the AEC those that should be translated and made generally available.

Although Brookhaven has virtually ceased to publish its own reports on a classified basis, such documents originating at other sites are still needed in some of the Laboratory's work, especially in the field of nuclear engineering. Therefore the Classified Library has continued to acquire classified reports, though on a smaller scale than formerly. Approximately 800 titles were received during the current report period; 340 were declassified and released to the Research Library. Since the Classified Library is the only remaining permanent exclusion area on the site, for security reasons the majority of classified reports have been recalled from other areas and limitations have been placed on the charging out of such documents. This has resulted in considerably increased use of the Library's facilities, in spite of the smaller number of classified holdings. The usual annual inventory of secret research and development reports was completed in February; all documents in this category have been accounted for during the past three years.

The Publication Group has edited and otherwise processed for issuance almost 60 major technical reports, including the *Proceedings of the Inter-*

*American Symposium on the Peaceful Application of Nuclear Energy*, held here May 13-17, 1957. This document, consisting of more than 70 technical papers and speeches presented by U.S. and Latin-American scientists and administrators, and the accompanying discussions, is being published in both English and Spanish with the report number TID-7554.

Another aspect of technical information services is the processing of all technical manuscripts originating from Brookhaven-sponsored research for publication in the scientific and technical journals. During the year 473 such papers (including published abstracts of speeches) have been processed, as compared with 401 for fiscal 1957, 393 for fiscal 1956, and 418 for fiscal 1955.

The distribution lists for the Laboratory's unclassified reports continue to grow. Standard distributions are made as prescribed by the AEC; in addition, all bona fide requests from within the national atomic energy project and from cooperating government agencies and universities are honored. Now that many foreign countries are developing national atomic energy programs, it has been possible to establish a number of fruitful exchange arrangements. Of special note is an extensive exchange of information, mainly in the field of high energy physics, with the Joint Institute for Nuclear Research in Moscow and that institute's high energy laboratory at Dubna. The benefits of this type of free and spontaneous exchange of scientific and technological information are apparent, from both the technical and political viewpoints.

#### PHOTOGRAPHY AND DUPLICATING

The activities of the photography group increased generally, as is shown in Table 15.

By dividing a high-ceilinged area into two stories, 480 square feet of space were added to the

Table 15

	Fiscal '58	Fiscal '57	Fiscal '56
Photographs	7,198	6,322	4,552
Photomicrographs	1,732	1,306	1,770
Lantern slides	5,946	6,251	6,283
Prints	57,255	41,583	36,351
Photostats, Xerox	22,127	23,627	19,925
Ozalid prints	429,625	295,517	332,451
Recording film processed	16.4 mi.	22.5 mi.	14.6 mi.

Table 16

	Fiscal '58	Fiscal '57	Fiscal '56
Offset pages	6,361,665	5,163,705	4,574,215
Offset negatives	8,738	7,034	5,479
Mimeo impressions	1,228,386	1,633,468	1,517,156
Stencils cut	728	1,011	1,497
Stencils run	11,424	11,266	10,084
Sheets collated and bound	2,388,700	1,951,525	1,637,145
Report copies issued	65,715	71,075	67,155

second floor of the photography building. This permitted a much needed expansion of the dark room area. The problem of space has been somewhat alleviated, and the efficiency of the operation has been definitely improved.

A 16 to 70-mm continuous film processing machine to handle the output from the bubble chambers has been placed on order.

Because of the increase in report printing, the duplicating group placed the Fotosetter on 3-shift operation which necessitated the hiring of two additional operators. This machine is well suited to the type of composition required at the Laboratory and has effected a reduction in costs. A Multigraph single-sheet collator is being used in the assembly of booklets. Production figures for the period are given in Table 16.

The illustration group made 2809 technical drawings for publications or for slides and was active in preparing exhibit material.

Preparations for the second Geneva Conference produced a peak of activity throughout the Division.

## Administration and Operations

The management operations reviewed in this section have been carried out in a manner designed to facilitate and encourage the research activities of the Laboratory's scientific staff.

### PERSONNEL

#### Scientific Staff

For the first time in several years there was a noticeable increase (6%) in the size of the regular scientific staff. The composition of the total scientific staff on May 31, 1958, is compared in Table 1 with that on the same date in 1957. As in the past, the turnover in regular staff was high. During the year there were 56 additions and 38 terminations. The increase in the number of Assistant Scientists is attributed to the fact that this appointment category was established only

three years ago. Its size relative to the total regular staff is now approaching equilibrium.

The number of salaried visitors increased by more than 10% for the third successive year. This increase is again traceable primarily to an increased number of salaried Research Associates. Of the 52 salaried and nonsalaried individuals in this category, 19 are foreign scientists. These appointments are similar to postdoctoral research fellowships and are limited in term. During the year 30 Research Associates were appointed, and 21 previous appointees completed their work and terminated.

The number of nonsalaried visitors holding appointments to the staff continued a trend established several years ago. The increase from 240 in 1957 to 272 in 1958 is entirely in the category of visitors who work at the Laboratory intermittently for one or more days at a time.

Table 1  
Scientific Staff on May 31, 1957 and 1958

	Visitors					
	Regular staff		Salaried		No compensation	
	1958	1957	1958	1957	1958	1957
<b>By appointment category</b>						
Senior Scientist	40	39	2	2	13	12
Scientist	87	82	3	2	98	89
Associate Scientist	104	103	6	2	88	65
Assistant Scientist	51	40	1	1	17	24
Research Associate	—	—	43	38	9	5
Junior Scientist	19	19	1	1	6	9
Junior Research Associate	—	—	9	10	35	28
Research Assistant	0	0	0	1	6	8
Total	301	283	65	57	272*	240**
<b>By academic degree</b>						
PhD or MD	175	174	51	44	190	155
Master	40	35	7	6	39	35
Bachelor	83	71	7	7	41	45
No degree	3	3	—	—	2	5

\*215 of these appointees worked intermittently at BNL.

\*\*177 of these appointees worked intermittently at BNL.

Table 2

Classification of Visiting Scientists Participating in BNL Program  
for Period of One Month or More, June 1, 1957 – May 31, 1958

	Guests and salaried visitors				1957 Summer program		Total	
	More than 3 months		Less than 3 months		Salaried	Guest	Individuals	Institutions
	Salaried	Guest	Salaried	Guest	Salaried	Guest		
University staff	12	21	3	25	47	82*	190	95
Thesis students	14	12	0	7	1	8	42	17
Student Research Assistants	2	0	3	1	87	33	126	45
Total	28	33	6	33	135	123	358	116 different
Industry	1	26	0	9	—	1	37	23
Other institutions	3	17	1	25	5	18	69	48
Total	32	76	7	67	140	142	464	187 different

\*Includes 30 individuals participating in Summer Institute in Nuclear Science and Engineering.

May 31 was selected as an appropriate date for Table 1, in order to exclude from the statistics individuals at the Laboratory only during the summer. Similarly, the dates used in Table 2 were selected in order to include participants in only one summer program, that of 1957. Dates coincident with the fiscal year would have spanned portions of two summer programs.

Table 2 shows the number of visiting scientists who worked at the Laboratory for a cumulative time of one month or more between June 1, 1957, and May 31, 1958. During this period, 358 individuals from 116 colleges and universities participated in research at Brookhaven. This compares with 303 individuals from 101 institutions last year, and 280 individuals from 86 institutions for the similar period two years ago. There were 106 individuals from industrial and other organizations, such as hospitals and government agencies, as compared with 93 last year and 85 two years ago. Graduate students engaged in doctoral investigations numbered 42 as compared with 37 last year and 34 two years ago. Sixteen students completed the research required for their degrees during the year. The numbers of university staff and students at the Laboratory during the summer represent a significant fraction of all visitors here for more than one month. For this reason, summer participants are indicated separately in Table 2.

Scientists and students who worked at the Laboratory for a cumulative time of less than one month are not included in Table 2. There were 109 such individuals here this year as compared with 90 last year and 85 two years ago.

A significant change in the composition of the scientific staff concerns participation by foreign scientists (Table 3). During the fiscal year ending June 30, 1956, 60 foreign visitors worked at the Laboratory. During the year just ended, there were twice that number. These scientists come from many countries (25 this year), and they work in almost every department of the Laboratory. In this respect, the Applied Mathematics Division is represented this year for the first time. In most instances foreign visitors receive their financial support from their own institutions or governments; several receive grants-in-aid from philanthropic organizations or U.S. Government agencies other than the Atomic Energy Commission; and about 35 are salaried, including 18 Research Associates. The presence of foreign scientists working cooperatively with others at the Laboratory continues to be stimulating to all individuals concerned.

Table 4 lists the man-days of service rendered by consultants. Although the number of consulting contracts in effect has increased, the quantitative use of consulting services has steadily declined since 1954.

### Summer Program for 1958

Arrangements have been made for 76 faculty members from 45 educational institutions to work at the Laboratory for a maximum of three months during the summer of 1958. In addition, 17 scientists and engineers from 13 other institutions and organizations will participate in the summer program at Brookhaven.

The formal summer student program has been continued for the seventh consecutive year. In January, announcements were sent to about 140 schools, most of which are located in the north-

eastern region of the United States. Applications were received from 69 graduate and 352 undergraduate students from 127 colleges and universities. Of the 79 appointments offered, 61 have been accepted by 20 graduate and 41 undergraduate students representing 40 institutions. In the student program, 25 fewer appointments were offered this year than last year because of the forced shutdown of the Cosmotron and the planned move of the Medical Department into the new Medical Research Center.

Also here for a portion of the summer are 18 Radiological Physics Fellows who have completed

Table 3

Foreign Visitors,\* July 1, 1957 – June 30, 1958,  
Appointed to Staff for Various Periods (Weeks to a Year)

Country	Field							Total for previous year (7/1/56-6/30/57)	
	High Energy and Theoretical Physics	Other Physics	Chemistry	Biology	Engineering	Mathematics	Medicine		
Germany	1	4	6	3		1	2	17	12
Britain	5	3	6			1	1	16	10
Japan	2	3	4	1	1			11	9
Canada		4	1	3			2	10	5
France	3	3	3		1			10	5
India	1	2	4	1	1			9	4
China	5		1	1	1			8	7
Italy	2	1	1		1		1	6	3
Israel		2	2	1				5	4
Turkey	1	1	2				1	5	1
Belgium		1			1		1	3	7
Netherlands		1	1					2	3
South Africa			2					2	2
Argentina	1				1			2	1
Australia	1			1				2	1
Sweden		1	1					2	1
Yugoslavia	1			1				2	1
Norway		1			1			1	2
Pakistan				1				1	1
Thailand				1				1	1
Finland					1			1	0
Indonesia			1					1	0
Ireland	1							1	0
Philippines						1	1	1	0
Scotland				1				1	0
Egypt								0	2
Spain								0	2
Chile								0	1
Greece								0	1
Total	24	27	35	15	8	3	8	120	86

\*This table does not include 28 foreign visitors here during the summer of 1958.

Table 4  
Consultants' Services

	1958	1957	1956
Total contracts in effect June 30	93	80	72
No. consultants used	46	35	41
No. man-days of service	308	358	443

the first part of their training at the University of Rochester; 17 additional students, most of whom were brought to the Laboratory by faculty members to work as their research assistants; and one Junior Research Associate working on his doctoral thesis.

The scientific staff will thus be increased by 190 additional workers. In addition, 30 faculty members will spend one week at the Laboratory in the Summer Institute in Nuclear Science and Engineering.

#### Personnel Management

**Employment.** The total number of employees at Brookhaven National Laboratory on June 30, 1958, excluding temporary appointees, research collaborators, and guests, was 1828. This represents a net increase of 132 staff members during fiscal 1958 – one of the largest increases in the Laboratory staff in several years. The majority of the new employees were added to scientific departments. For example, in the Physics Department there were 13 additions; in both the Nuclear Engineering and Accelerator Development Departments, 25. The ratio of one scientist to four nonscientists at the Laboratory remained unchanged from that for the previous year. A comparison of employment statistics for fiscal 1957 and 1958 is given in Table 5.

A more favorable employment market and a substantial increase in advertising combined to permit the Laboratory a higher degree of selectivity in employing new staff members than has heretofore been practicable.

**Labor Relations.** The two-year labor agreement with the Directly Affiliated Labor Union No. 24426, AFL-CIO, covering approximately 385 maintenance, shop, and fire-fighting employees, was in effect during fiscal 1958 and will not terminate until December 31, 1958. Reopening the contract at the end of the first year of its term, for the limited purpose of negotiation of

wages and job evaluation procedures, brought about extended bargaining sessions.

The negotiation of a second one-year labor contract with the Oil, Chemical, and Atomic Workers International Union, covering 17 pile operators, also required many bargaining sessions.

A two-year labor contract with the International Guards Union of America, covering 23 Laboratory policemen, was in effect during fiscal 1958; its termination date is February 28, 1959.

**Salary Administration.** A major achievement in personnel management during fiscal 1958 was the completion of a salary administration survey. A comprehensive study was made of all professional, technical, administrative, and supervisory positions, and position descriptions and evaluations were revised or established as necessary. An extensive salary survey was conducted, and revised salary ranges were promulgated. The results are already evident in better work performance and improved employee morale.

**Training and Publications.** During the fiscal year a revised Salary and Wage Manual was prepared for distribution to supervisors. In addition, a Supervisors' Personnel Manual was prepared to provide a ready reference for supervisory personnel on Laboratory personnel policies and procedures. Work is well advanced on a new Employees' Handbook for distribution to all employees.

**Employee Activities.** The Brookhaven Employees Recreation Association (BERA) not only continued its successful programs but entered into new activities which were enthusiastically received. A summer program of athletics and swimming for children of employees was very well attended, and the first annual picnic for employees, their families, and guests was attended by 3000 men, women, and children.

The recreation building on York Lane in the apartment area has been improved. BERA has completely furnished the common room and will employ an attendant to oversee the activities in the building. As in the past, the building will serve as a recreation center for both on-site and off-site employees and as a general meeting place for BERA activities and groups. In view of increased tennis activity, the tennis courts were resurfaced.

**Employee Benefit Plans.** In continuance of the Laboratory's policy of providing employees with up-to-date benefits, the Accidental Death feature

Table 5

## Employment Statistics

Turnover data	Number	Fiscal 1958		Fiscal 1957		
		Annual rate (%)	Average monthly rate (%)	Number	Annual rate (%)	Average monthly rate (%)
Accessions	329	18.76	1.56	348	21.17	1.76
Separations (turnover)	197	10.96	0.91	250	15.26	1.27
Net accessions	132	7.53	0.63	98	5.93	0.49
Replacements (net turnover)	192	10.67	0.89	233	14.20	1.18

\*Not including 139 temporary appointees.

\*\*Not including 144 temporary appointees.

of the Group Life Insurance Plan was extended from "at work" coverage to 24-hr coverage.

In addition, provision was made for extension of the Laboratory's Supplementary Life Insurance program to all permanent employees. As a result, participation in this phase of employee benefits increased from 35% to 57% of all employees.

### SECURITY AND PLANT PROTECTION

The Security and Plant Protection Division functions as both a security and a service organization. As a security organization, it administers and enforces the AEC and Laboratory regulations pertaining to the safeguarding of restricted data and the clearance of Laboratory personnel and visitors. As a service organization, it provides for the protection of life and property through the activities of the Police and Fire Groups.

While the importance of security at the Laboratory has not diminished, the scope of the security program continues to contract by reason of the Atomic Energy Commission's declassification program and the consequent reduction of classified work at the Laboratory. A small repository of classified documents in the Nuclear Engineering building is the sole restricted area on the site. On the other hand, there has been a substantial increase in the number of guests, visitors, and subcontractor employees on the site, all requiring some measure of attention from Security personnel.

The increase in unclassified research, the greater utilization of existing buildings, and the wider variety of research investigations have combined

to bring about an expansion in the plant protection program. The problems arising from these conditions have, however, been successfully resolved in each instance.

An indication of the growth in activities is provided in Table 6, which shows the number of admissions of nonstaff personnel to the site.

One fire of consequence occurred during the fiscal year. This fire was effectively controlled and confined to the experimental area in which it originated. Although serious, it did not require utilization of the Fire Group's full resources, nor of the Emergency Disaster Teams. The activity of the Fire Group is shown in the tabulation below.

	1958	1957	1956
Responses on site	113	109	80
Responses off site	2	2	5
No response - investigation	14	14	11
Total	129	125	96

### ARCHITECTURAL PLANNING

The Architectural Planning Division continued its collaborative efforts with the Brookhaven Area Manager's Office and their Architect-Engineers in connection with the construction of the Medical Research Center. Similar assistance has been rendered in connection with two projects currently under AEC construction contract, namely, an addition to the cyclotron-Van de Graaff building to house the 18-in. cyclotron and the hot laundry and reclamation facility.

Table 6  
Admissions to the Site

	Fiscal 1958	Fiscal 1957	Fiscal 1956
Regular visitors*	30,555	28,287	22,507
Conference visitors	4,687	2,760	2,452
Alien visitors	1,931	1,969	1,179
Subcontractor employees	21,273	20,747	17,610
Total	58,446	53,763	43,748

\*Includes visitors during open house days.

Another type of joint construction effort concerned the Cosmotron target area project. This Division was assigned complete responsibility for the design and supervision of a major portion of required modifications to the existing building, all site work adjacent to the new addition, and all utility revisions and extensions (water, steam, and underground electric mains). Most of this work, which will have a total value of more than \$150,000, is now in progress, and completion is planned to coincide with that of target area construction. In addition, the Division produced the engineering design of the heavy concrete shielding blocks for the Cosmotron.

In collaboration with the Nuclear Engineering Department, the Division provided engineering design and is now supervising construction of the hot metallurgical facility in the Hot Laboratory building. This facility is basically a heavily shielded cell with extensive, remotely operated equipment for metallurgical examination of highly radioactive samples. The cost of its construction and installed equipment will be more than \$350,000. The work is scheduled for essential completion late in 1958.

During the year many other jobs, with a total value of more than \$700,000, were undertaken that required working drawings and specifications, contract administration, and field supervision by this Division. The principal projects under this heading included

Feeder substation in Medical Research Center for Biology underground electric mains	\$ 54,000
Alterations in building for Solid State Physics	35,000
Building addition for Critical Assembly Laboratories	31,000
Pulsed liner for Cosmotron injector	23,000

Two-loop shed addition to works area building	68,000
Research Library extension (in design)	60,000
Reactor projects including ground drainage contamination control, staff shop relocation, reactor works area, and east wing basement	75,000
Biology projects including new high pressure steam connection and extension to west greenhouse	37,000
Modifications and additions to the Metallurgical and Hot Laboratories and to the Nuclear Engineering and Chemistry buildings	57,000
Power supplies, Cosmotron	120,000
Apartment heating	125,000

### PLANT MAINTENANCE

The Plant Maintenance Division is responsible for the operation of all plant utilities, the maintenance of all buildings, roads and grounds, and the provision of communications, transportation, travel, and housing services to all segments of the Laboratory. Other services include rigging and moving, maintenance of motor vehicles, janitorial service, and model-making and carpentry in connection with scientific projects.

A comparative breakdown of the utilization of manpower within the Plant Maintenance Division in fiscal 1957 and 1958 is shown in Table 7. A comparative analysis of major costs involved in the operation and maintenance of plant utilities is presented in Table 8. Only general and administrative costs are included in this table. Special maintenance work was accomplished during the past year on laboratory, service, and administrative buildings. Included in this program was the continuing rehabilitation of housing units. The major exterior maintenance items consisted of waterproofing five buildings, reroofing seven buildings, and shingling dormitory and housing units in lieu of painting. Interior maintenance of special nature included the replacement of deteriorated utilities, repainting, restoration of floors, and major carpentry repairs. The projects were carried on in 21 buildings embracing varied centers of activity throughout the Laboratory.

### SUPPLY, MATERIEL, AND INVENTORY MANAGEMENT

The Supply and Materiel Division is responsible for the storage and issue of approximately 22,000

Table 7  
Manpower Utilization

Type of work	Fiscal 1958		Fiscal 1957	
	Productive man-years	Percent of total work	Productive man-years	Percent of total work
Plant utilities operation	53	23	51	22
Building janitor service	29	12	24	10
Decontamination and hot laundry	11	5	9	4
Total plant operations	93	40	84	36
Communications service	13	6	14	6
Transportation, housing, and staff services	17	8	20	9
Total staff services	30	14	34	15
Special services for others	36	16	34	15
Facility improvements	3	1	3	1
Total special services	39	17	37	16
General repairs and maintenance	44	20	60	26
Special long-term maintenance	21	9	16	7
Total maintenance	65	29	76	33

Table 8  
Costs of Supplies, Materials, and Purchased Labor

	Fiscal 1958	Fiscal 1957
Plant utilities operation		
Coal and fuel oil	\$ 280,156	\$ 256,365
Electricity	685,973	752,879
General supplies	15,616	23,680
Building janitor supplies	16,773	13,086
Decontamination and hot laundry supplies	23,463	24,132
Total plant operations	1,021,881	1,070,142
Staff services		
Telephone, teletype, and mail	197,586	160,117
Transportation (gasoline)	15,707	14,678
Housing and cafeteria supplies	74,667	36,334
Total staff services	287,960	211,129
Maintenance		
General material and purchased labor	136,734	109,603
Special materials and purchased labor	542,507	219,339
Total maintenance	679,241	328,942
Grand Total	\$1,989,082	\$1,610,213

Table 9  
Inventory Changes

Type of inventory	Fiscal Year		
	1958	1957	1956
Active inventory			
Opening active inventory July 1	\$293,056	\$261,146	\$274,574
Net of purchases, stores issues, adjustments, additions to inventory, and transfers to excess	+24,850	+31,910	-13,428
Closing active inventory June 30	317,906	293,056	261,146
No. of months' investment in active inventory June 30	3.3	3.3	3.8
Stand-by inventory			
Opening stand-by inventory July 1	3,402	0	
Net of purchases, issues to active, and adjustments	-3,402	3,402	
Closing stand-by inventory June 30	0	3,402	
Excess inventory			
Opening excess inventory July 1	0	4,327	45,026
Net of transfers from active inventory, dispositions, and adjustments	0	-4,327	-40,699
Closing excess inventory June 30	0	0	4,327
Total inventory			
Opening total inventory July 1	296,457	265,473	319,600
Net of purchases, stores issues, adjustments, transfers to excess, and dispositions	+21,448	+30,984	-54,127
Closing total inventory June 30	\$317,905	\$296,458	\$265,473

inventory items. During the year several items previously acquired by direct purchase were added to the inventory account. Approximately 16,000 items are issued only on signed requisitions and are subject to daily machine records control.

Probably the most significant achievement during the fiscal year in the area of supply has been the installation of an effective yet inexpensive method of dispensing approximately 6000 low cost, common usage inventory items, such as nuts, bolts, screws, resistors, and other high volume, low unit cost supplies. These items are now stocked and issued without formal requisitions through a centrally located self-service storeroom.

Despite the increase in the volume of stores issues and the other services now provided by this Division, there has been no increase in personnel, and the volume of space allotted to these functions has been significantly decreased. A comparison of personnel strength and functions performed in 1958 and in 1954 indicates that the same functions are now being performed by 12 fewer employees.

The utilization of committees composed of ultimate users of inventory items to consult with and advise inventory management personnel on the adequacy and balance of the inventory has increased substantially during the fiscal year. These committees provide invaluable aid in the task of maintaining an inventory which is at minimum level yet responsive to the supply needs of the research staff. Although Table 9 reflects a small increase in inventory investment in fiscal 1958 as compared to previous years, it should be noted that the number of months' investment in active inventory remained constant with the prior year's low of 3.3 months. This indicates an inventory turnover of approximately 3½ times per year.

In the maintenance of residential and dormitory furniture and furnishings, the practice of refinishing along with replacement has resulted in substantial savings.

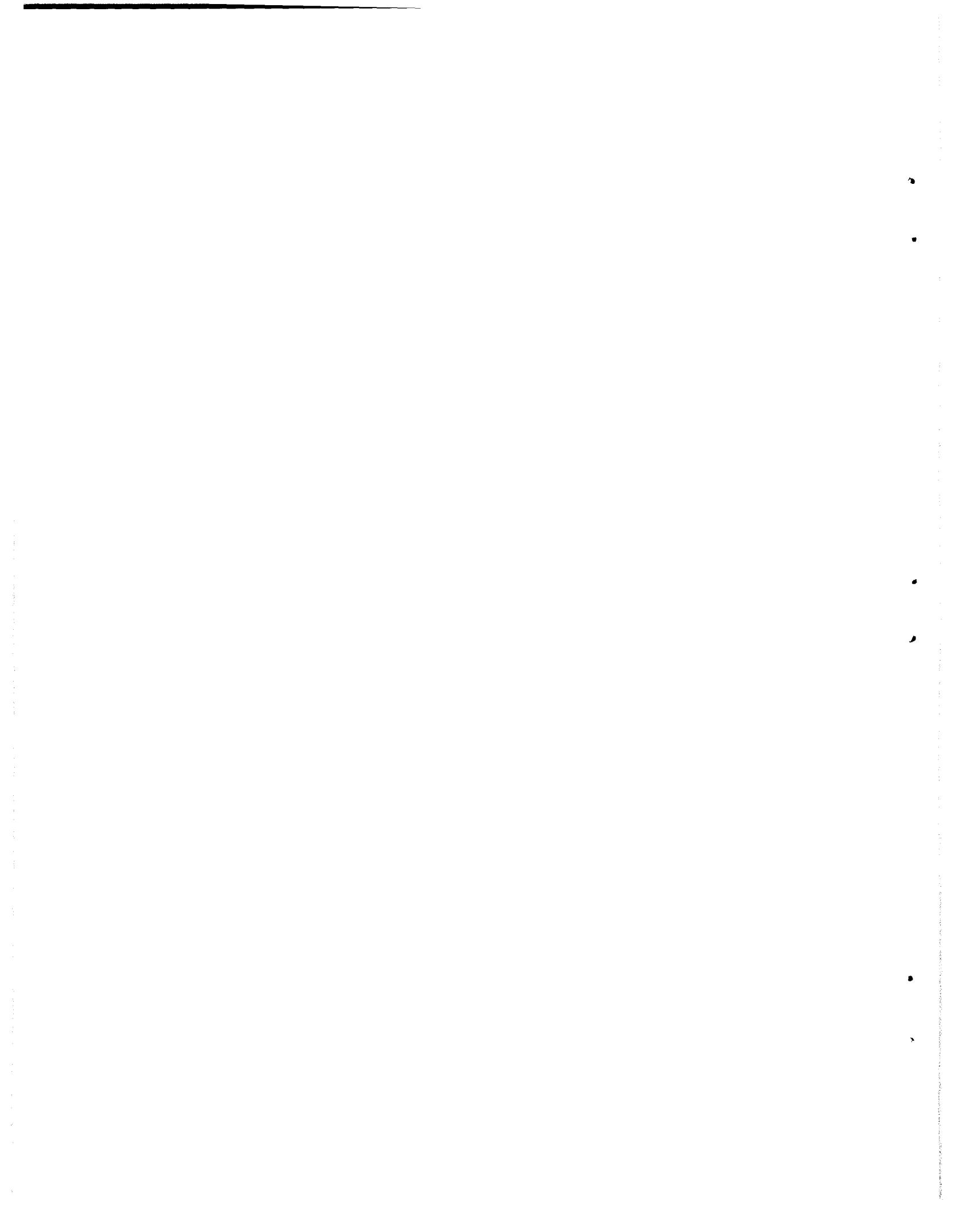
The Supply and Materiel Division complements the Purchasing Group in the implementation of the petty cash purchase program by making daily pickups in nearby communities. It also operates a receiving and distributing facility at the site of

the Alternating Gradient Synchrotron, which has expedited the receipt and delivery of supply items to the ultimate user in the AGS project.

#### **PURCHASING**

For the past fiscal year the total dollar volume of purchase orders, excluding research, develop-

ment, and construction contracts, was \$5,619,687 as compared with \$4,666,372 in the previous year, an increase of 20%. The Laboratory paid \$40,171 for 57 purchases from government lists of excess material and equipment which had an original acquisition cost of \$373,477. One sale of excess material and equipment was conducted which resulted in a return of \$2439.



## Appendix A

### UNCLASSIFIED PUBLICATIONS, JULY 1, 1957 - JUNE 30, 1958

This list includes official Laboratory publications, abstracts of papers which were or will be presented at scientific meetings, and publications by staff members, consultants, and guests. All these listings result from work done at the Laboratory; they were submitted during the review period.\* Abstracts are indicated by (A); letters to the editor, (L); and notes, (N). Acceptance for future publication is designated by (In press).

#### GENERAL PUBLICATIONS

Annual Report, July 1, 1957. BNL 462 (AS-11).  
Quarterly Progress Reports:  
July 1 - September 30, 1957. BNL 473 (S-38).  
October 1 - December 31, 1957. BNL 484 (S-40).  
January 1 - March 31, 1958. BNL 502 (S-42).  
April 1 - June 30, 1958. BNL 515 (S-44).  
Quarterly Progress Reports, Nuclear Engineering Department:  
February 15 - December 31, 1956, BNL 434 (S-34).  
January 3 - April 30, 1957, BNL 472 (S-37).  
May 1 - September 30, 1957, BNL 477 (S-39).  
October 1 - December 31, 1957, BNL 491 (S-41).  
January 1 - March 31, 1958, BNL 506 (S-43).  
Conference Reports:  
Brookhaven Symposia in Biology No. 10. *Homeostatic Mechanisms*. BNL 474 (C-25).  
*Proceedings of the French-American Conference on Graphite Reactors*. BNL 489 (C-27).  
Weekly Bulletin 10, No. 50-52; 11, No. 1-50.  
Weekly Selected Reading List 10, No. 18-52; 11, No. 1-17.  
Miscellaneous:  
*Bibliography on the Effects of Ionizing Radiation on Plants, 1896-1955*. A.H. SPARROW, J.P. BINNINGTON, AND V. POND. BNL 504 (L-103), July 1958.

#### STAFF PUBLICATIONS AND ABSTRACTS

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\*Also included are those listings from the last Annual Report [BNL 462 (AS-11)] for which complete reference information was not then available.

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WARBURTON, E.K. (See Rose, H.J.)

WENESER, J. (See Church, E.L.)

WHITTEMORE, W.L. (See Bolze, E.M.)

WHITTEMORE, W.L. (See Andresen, A.)

WHITTEMORE, W.L. (See Cool, R.L.)

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ZIMMERMAN, R.L. (See Fox, J.D.)

ZIMMERMAN, R.L. (See Chrien, R.E.)

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### Reactor Division

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STOCK, E.V. Slowing down and diffusion lengths of  
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OSBORNE, C.L. (See Powell, R.W.)

PHELPS, J.P. (See Hendrie, J.M.)

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Aug. 1957 BNL 469 (T-105)  
Sept. 1957 BNL 475 (T-108)  
Oct. 1957 BNL 488 (T-113)  
Nov. 1957 BNL 480 (T-106)  
Dec. 1957 BNL 492 (T-114)  
Jan. 1958 BNL 495 (T-116)  
Feb. 1958 BNL 503 (T-120)  
Mar. 1958 BNL 507 (T-121)  
Apr. 1958 BNL 519 (T-127)  
May 1958 BNL 525 (T-130)  
June 1958 BNL 517 (T-126)

POWELL, R. (See Corngold, N. - Nuclear Engineering)

PRICE, G.A. (See Hendrie, J.M.)

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SEGEL, R.E. (See Kienle, P. - Instr. & Health Physics)

SEGEL, R.E., KANE, J.V. AND WILKINSON, D.H. Parity  
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WEINSTOCK, E.V. (See Hendrie, J.M.)

WEINSTOCK, E.V. (See Kane, J.V. - Physics)

## Appendix B

### OFFICERS AND SCIENTIFIC STAFF

Leland J. Haworth, *Director*  
Gerald F. Tape, *Deputy Director*  
William H. Fields, Jr., *Associate Director*

R. Christian Anderson, *Assistant Director*      Robert A. Patterson, *Assistant Director*  
Samuel M. Tucker, *Assistant to Director*

Leland J. Haworth, *Chairman*, Accelerator Development Department  
Milton E. Rose, *Head*, Applied Mathematics Division  
Howard J. Curtis, *Chairman*, Biology Department  
Richard W. Dodson, *Chairman*, Chemistry Department  
George B. Collins, *Chairman*, Cosmotron Department  
J.B.H. Kuper, *Chairman*, Instrumentation and Health Physics Department  
Gerald F. Tape, *Acting Head*, Mechanical Engineering Division  
Lee E. Farr, *Chairman*, Medical Department  
Clarke Williams, *Chairman*, Nuclear Engineering Department  
Samuel A. Goudsmit, *Chairman*, Physics Department  
Robert W. Powell, *Head*, Reactor Division

J. Georges Peter, *Director*, Architectural Planning  
H. Russell Cort, *Budget Officer*  
Lewis R. Burchill, *Controller*  
Charles F. Dunbar, *Legal Counsel*  
Edward A. Macy, \* *Personnel Officer*  
Joseph S. Washburne, *Personnel Manager*  
Frederick H. Williams, *Security Officer*

---

#### Accelerator Development Department

Leland J. Haworth, *Chairman*  
Roger R. Adams  
Richard A. Beth  
John W. Bittner  
John P. Blewett  
M. Hildred Blewett  
Hugh N. Brown  
Nicholas C. Christofilos\*  
Theodore N. Constant  
James G. Cottingham  
Gordon T. Danby

\*Terminated before June 30, 1958.

Charles E. Falk  
(*on leave to Atomic Energy Commission, Washington, D.C.*)  
Theodore Fishman  
Robert J. Gerrity  
Salvatore T. Giordano  
George K. Green  
Ralph K. Kassner  
John D. Kiesling  
William T. Link  
Robert J. McCracken  
Lowell McLean  
Thomas F. Madigan  
James P. Palmer  
Robert H. Phillips  
Martin Plotkin  
Eugene C. Raka

Raymond H. Rheaume  
Maxwell M. Small  
Julius Spiro  
Philip Stehle\*  
(*on leave from Univ. of Pittsburgh*)  
Arie Van Steenbergen

#### Applied Mathematics Division

Milton E. Rose, *Head*  
Bruno F. Dejon  
David L. Fox  
(*on leave from NYU*)  
Ray C. Makino  
Martin Milgram  
Alfred J. Mumford  
Stuart S. Rideout

Yoshio Shimamoto  
Peter M. Treuenfels

### Biology Department

Howard J. Curtis, *Chairman*  
Irvin C. Anderson  
(*postdoctoral appointment*)  
Leroy G. Augenstine  
John A. Bergeron  
James L. Brewbaker  
Phelps P. Crump  
Robin L. Cuany  
(*postdoctoral appointment*)  
Donald J. Fluke\*  
Rufus C. Fuller  
Earl B. Herr, Jr.\*  
(*postdoctoral appointment*)  
Werner Hirs  
James R. Innes  
James R. Klein  
Calvin F. Konzak\*  
Daniel E. Koshland, Jr.  
Marian E. Koshland  
Leo E. La Chance  
(*postdoctoral appointment*)  
Richard L. Latterell  
(*assigned from*  
*U.S. Public Health Service*)  
Myron Levine  
Harvey M. Levy  
(*postdoctoral appointment*)  
Paul Margolin  
(*assigned from*  
*U.S. Public Health Service*)  
Leo W. Mericle\*  
(*on leave from Michigan State Univ.*)  
Rae P. Mericle\*  
(*assigned from E. W. Sparrow Hospital*)  
Jonas W. Miller  
(*graduate student from Cornell Univ.*)  
Ronald F. Myers  
(*graduate student from*  
*Univ. of Connecticut*)  
Arth Nakornthap\*  
(*on leave from*  
*Kasetsart Univ., Thailand*)  
Leslie F. Nims  
Henry Quastler  
William J. Ray  
(*postdoctoral appointment*)  
Yoneo Sagawa\*  
(*postdoctoral appointment*)  
Leon Z. Saunders\*  
Erwin A. Schwinghamer  
Seymour Shapiro  
Oved Shiffriss\*  
(*on leave from*  
*Weizmann Inst. of Science, Israel*)

Edward C. Sisler  
(*postdoctoral appointment*)  
Harold H. Smith  
Arnold H. Sparrow  
Robert Steele  
Dale M. Steffensen  
Otto L. Stein  
(*on leave from Univ. of Minnesota*)  
Robert E. Swanson\*  
Paul A. Swenson\*  
(*on leave from Univ. of Minnesota*)  
Stephen T. Takats  
(*assigned from*  
*U.S. Public Health Service*)  
Donald E. Wimber  
(*on leave from Columbia Univ.*)  
Marilyn L. Wolfsberg  
(*on leave from NYU*)  
Philip S. Woods

### Chemistry Department

Richard W. Dodson, *Chairman*  
Augustine O. Allen  
Saadia Amiel  
(*postdoctoral appointment*)  
Elizabeth W. Baker  
Bahattin M. Baysal  
(*on leave from*  
*Univ. of Ankara, Turkey*)  
Envare A. Baysal  
(*on leave from*  
*Univ. of Ankara, Turkey*)  
Jacob Bigeleisen  
James M. Caffrey, Jr.  
(*assigned from The Texas Co.*)  
David R. Christman  
Lester M. Corliss  
James B. Cumming  
Raymond Davis, Jr.  
Adolf E. DeVries\*  
(*postdoctoral appointment*)  
Norman Elliott  
Simon Freed  
Gerhart Friedlander  
Lewis Friedman  
Rutherford J. Gettens\*  
(*assigned from Smithsonian Institution*)  
Barry M. Gordon\*  
(*postdoctoral appointment*)  
Floyd T. Gould  
(*on leave from Columbia Univ.*)  
James R. Grover  
(*postdoctoral appointment*)  
Walter C. Hamilton  
Garman Harbottle  
(*on leave as Guggenheim Fellow*  
*at Cambridge Univ., England*)  
Donald S. Harmer  
(*postdoctoral appointment*)  
Julius M. Hastings  
Wolfgang Henkes  
(*postdoctoral appointment*)

Erik H. Hogfeldt  
(*postdoctoral appointment*)  
Jerome Hudis  
Adolph P. Irska  
Seymour Katcoff  
(*on leave as Weizmann Fellow*  
*at Weizmann Inst. of Science, Israel*)  
Hosaku Kawahara\*  
(*on leave from*  
*Otaru College of Commerce, Japan*)  
Fritz S. Klein\*  
(*on leave from*  
*Weizmann Inst. of Science, Israel*)  
Alexander J. Kresge  
Esther Krikorian  
(*postdoctoral appointment*)  
Neil L. Lark  
(*graduate student from Cornell Univ.*)  
Norman N. Lichten  
(*on leave from Boston Univ.*)  
C. Alden Mead  
(*postdoctoral appointment*)  
Pasupati Mukerjee  
(*postdoctoral appointment*)  
James R. Nash  
(*assigned from Frankford Arsenal*)  
Tomota Nishi  
(*on leave from Kyoto Univ., Japan*)  
Elinor F. Norton  
Brian D. Pate  
Catherine T. Pawlowicz  
Morris L. Perlman  
Russell W. Pierce  
(*graduate student from Columbia Univ.*)  
Norbert T. Porile  
(*postdoctoral appointment*)  
Arthur M. Poskanzer  
(*postdoctoral appointment*)  
K. Narayano Rao  
(*graduate student from Boston Univ.*)  
Herbert E. Rauscher  
(*graduate student from Columbia Univ.*)  
Carol S. Redvanly  
Etienne G. Roth  
(*on leave from Commissariat  
à l'Energie Atomique, France*)  
Walter G. Rothschild  
(*on educational leave*)  
J. Keith Rowley  
William Rubinson  
William Salmre  
Ronald L. Sass  
(*postdoctoral appointment*)  
Derek P.N. Satchell  
(*postdoctoral appointment*)  
Edward V. Sayre  
Oliver A. Schaeffer  
George Scholes\*  
(*postdoctoral appointment*)  
Eric C. Schreiber  
(*assigned from*  
*Charles Pfizer and Co., Inc.*)

\*Terminated before June 30, 1958.

Harold A. Schwarz  
 Stanley Seltzer  
*(postdoctoral appointment)*  
 Patricia R. Sisler\*  
 Morris Slavin  
 James S. Smart  
*(on leave from  
U.S. Naval Ordnance Laboratory)*  
 Esther L. Sprenkel  
*(graduate student from  
Univ. of Rochester)*  
 Raymond W. Stoerner  
 Bulusu Suryanarayana  
*(postdoctoral appointment)*  
 James W. Sutherland  
*(postdoctoral appointment)*  
 Norman Sutin  
 Sydney O. Thompson  
 Ralph E. Weston, Jr.  
 Alfred P. Wolf  
 Max Wolfsberg  
 John Yang  
*(postdoctoral appointment)*  
 Josef Zähringer\*  
*(on leave from  
Univ. of Freiburg, West Germany)*

#### Cosmotron Department

George B. Collins, *Chairman*  
 Robert K. Adair  
 Mark Q. Barton  
 Henry Blumenfeld  
*(on leave from Duke Univ.)*  
 Theodore Bowen\*  
*(on leave from Princeton Univ.)*  
 Walter G. Chesnut\*  
*(postdoctoral appointment)*  
 William Chinowsky  
 Ernest D. Courant  
 Edward W. Dexter  
 Frederick R. Eisler  
 Cyril Henderson  
 David A. Hill  
*(on leave from MIT)*  
 Lawrence B. Leipuner  
 Isador J. Livant  
 Robert A. Loper\*  
 William H. Moore, Jr.  
 Roger B. Perkins  
*(graduate student from Princeton Univ.)*  
 Albert G. Prodell  
*(on leave from Barnard College)*  
 Edwin J. Rogers  
 John H. Scandrett\*  
*(graduate student from  
Univ. of Wisconsin)*  
 Melvin Schwartz\*  
 Lyle W. Smith  
 Rudolph Sternheimer

\*Terminated before June 30, 1958.

Clifford E. Swartz  
 Arthur Tranis  
 Clarence M. Turner  
 William D. Walker  
*(on leave from Univ. of Wisconsin)*  
 Albert Werbrouck  
*(graduate student from Princeton Univ.)*  
 Luke C.L. Yuan

#### Instrumentation and Health Physics Department

J.B.H. Kuper, *Chairman*  
 Richard H. Boutelle  
 Arland L. Carsten  
 Robert L. Chase  
 Frederick P. Cowan  
 Jack S. Cubert\*  
 Carl H. Distenfeld  
 Emile Donneaux\*  
*(assigned from  
Institut Interuniversitaire, Belgium)*

Edward H. Foster  
 Lee Gemmell  
 Thomas M. Gerusky  
 Martin H. Graham\*  
 John S. Handloser  
 Mortimer B. Heller\*  
 William A. Higinbotham  
 Roland P. Kenshaft  
 Paul Kienle  
*(on leave from Technische Hochschule,  
Munich, West Germany)*  
 Clifford J. Konnerth  
 Charles B. Meinholt  
 Gabriel L. Miller  
 Thomas D. Murphy  
 Casimir Z. Nawrocki  
 Leigh F. Phillips  
 David W. Potter  
 Gwendolyn S. Prodell\*  
 Ernest A. Rainey  
 Seymour Rankowitz  
 Francis X. Rizzo  
 Seymour N. Rottenberg  
 George E. Schwender  
 Irving A. Singer  
 Maynard E. Smith  
 Robert J. Spinrad  
 Raymond W. Stong  
 Robert M. Sugarman  
 Isaac Van der Hoven\*  
*(assigned from U.S. Weather Bureau)*  
 Jerome Weiss  
*(on leave as Fulbright Fellow at  
Univ. of Edinburgh, Scotland)*

#### Mechanical Engineering Division

Gerald F. Tape, *Acting Head*  
 Robert D. Baldwin, Jr.

Vernon J. Buchanan  
 Burton Z. Chertok  
 Basil De Vito  
 Charles L. Gould  
 John J. Grisoli  
 Kenneth C. Hoffman  
 Donald W. Huszagh  
 David D. Jacobus  
*(on leave to Harvard Univ.)*  
 Jack E. Jensen  
 Michael B. Karelitz  
 David A. Kassner  
 Calman Lasky  
 Walter W. Merkle  
 Kurt F. Minati  
 Irving J. Polk  
 Oliver S. Reading  
 Everett J. Rutan  
 Albert P. Schlafke, Jr.  
 Edward E. Shelton  
 William G. Walker

#### Medical Department

Lee E. Farr, *Chairman*  
 Howard J. Bagnall  
*(postdoctoral appointment)*  
 Albert J. Bertinchamps  
*(postdoctoral appointment)*  
 Victor P. Bond  
 Donald C. Borg  
 Robert W. Christie\*  
 Stanton H. Cohn  
 Robert A. Conard, Jr.  
 George C. Cotzias  
 Eugene P. Cronkite  
 Lewis K. Dahl  
 Roger C. DeMeutter  
*(postdoctoral appointment)*  
 Ruth M. Drew  
 Otho D. Easterday  
 Theodore M. Fliedner  
*(postdoctoral appointment)*  
 Conrad T.O. Fong  
 Lewis I. Gidez\*  
 Mary B. Hagamen  
 Lawrence V. Hankes  
 Mary M. Hawrisiak  
*(graduate student from  
Johns Hopkins Univ.)*  
 Allen R. Hennes\*  
 Walter L. Hughes  
 Werner E. Kahle  
*(postdoctoral appointment)*  
 Paul G. LeFevre  
 Stuart W. Lippincott  
 Robert A. Love  
 Myles Maxfield  
 Gerald F. McGinniss  
*(postdoctoral appointment)*  
 Robert B. Painter

Victor Perman  
(postdoctoral appointment)  
Edwin A. Popenoe  
James S. Robertson  
Joseph R. Rubini  
(postdoctoral appointment)  
Claire J. Shellabarger  
Walton W. Shreeve  
Lawrence Silver\*  
F. Marrott Sinex\*  
Dale K. Sorenson  
(postdoctoral appointment)  
Elmer E. Stickley  
Richard D. Stoner  
Geronimo Terres, Jr.  
Donald D. Van Slyke  
(assigned from Eli Lilly and Co.)  
Donald A. Willigan\*  
(on leave from Univ. of Minnesota)  
William Wolins

#### Nuclear Engineering Department

Clarke Williams, *Chairman*  
George Adler  
Seymour Adler\*  
Clemens Auerbach  
Theodore Auerbach  
David S. Ballantine  
Malcolm Basche\*  
Jacques Bernot\*  
(assigned from Commissariat  
à l'Energie Atomique, France)  
Fritz Bloch  
Romeo G. Bourdeau\*  
Merwyn B. Brodsky  
Joseph S. Bryner  
Catello Cesarano  
(assigned from Centro  
Informazioni Studi Esperienze, Italy)  
Jack Chernick  
Joe G. Y. Chow  
Noel R. Corngold  
Robert F. Doering  
Kenneth W. Downes  
Orrington E. Dwyer  
James J. Egan  
Allen Eshaya  
Harmon L. Finston  
Aaron J. Friedland  
(graduate student from Columbia Univ.)  
William S. Ginell  
Althea Glines  
Irwin Goldberg  
David M. Graham\*  
(assigned from Babcock and Wilcox Co.)  
Leon Green\*  
(assigned from  
United Engineers and Constructors Corp.)

David H. Gurinsky  
Loranus P. Hatch  
Joseph M. Hendrie  
Eric Hellstrand\*  
(assigned from AB Atomenergi, Sweden)  
Raymond J. Heus  
Frank B. Hill  
Frederick L. Horn  
Robert J. Isler  
William T. Johnsen  
(assigned from Babcock and Wilcox Co.)  
Richard Johnson  
Sheldon Kalish  
Otto F. Kammerer  
Irving Kaplan\*  
Herbert M. Katz  
William F. Kenney  
Carl J. Klamut  
George H. Knepple\*  
(graduate student from Pratt Inst.)  
Herbert J.C. Kouts  
Otto A. Kuhl  
Lawrence E. Kukacka, Jr.  
William E. McNulty  
Walter Majkowski\*  
(assigned from  
Grumman Aircraft Engineering Corp.)  
Bernard Manowitz  
David B. Marsland\*  
(graduate student from Cornell Univ.)  
Frank D. Maslan  
Jean Maurin\*  
(assigned from Compagnie  
Française de Raffinage, France)  
Donald J. Metz  
Robert A. Meyer  
Francis T. Miles  
Terukatsu Miyauchi  
(on leave from Univ. of California)  
Alexander Murrenhoff\*  
(on leave from Bonn Inst. for  
Physical Chem., West Germany)  
Donald Neil\*  
(graduate student from RPI)  
Leonard Newman  
Granville M. Olds  
(assigned from Babcock and Wilcox Co.)  
Sophie Oleksa  
Betty Perkins\*  
James P. Phelps  
Carl Pierce  
James R. Powell, Jr.  
Glenn A. Price  
Chad J. Raseman  
William H. Regan, Jr.  
David M. Richman  
Arthur E. Roswell  
Wolfgang Rothenstein  
(postdoctoral appointment)  
Patrick Russell  
(assigned from Babcock and Wilcox Co.)  
Jerome Sadofsky

Francis J. Salzano  
César A. Sastre  
(postdoctoral appointment)  
Kenzo Sato

(assigned from Mitsubishi  
Shipbuilding Engrg. Co., Japan)  
Clifford H. Scarlett  
Donald G. Schweitzer  
Rudolph Sher  
John L. Speirs  
Louis G. Stang, Jr.  
Eugene Starr  
Meyer Steinberg  
Gerald Strickland  
Herbert Susskind  
Walter D. Tucker  
Charles H. Waide  
John R. Weeks  
Eugene V. Weinstock  
Henry H. Windsor  
Edward Wirsing, Jr.  
Richard H. Wiswall, Jr.  
Samuel A. Zwickler

#### Physics Department

Samuel A. Goudsmit, *Chairman*  
David E. Alburger  
Frederick Ayer II  
Charles P. Baker  
Saul Barshay  
(postdoctoral appointment)  
Adam M. Bincer  
(postdoctoral appointment)  
Marietta E. Blau  
Stewart D. Bloom\*  
Ernest M. Bolze  
Morton K. Brussel  
(postdoctoral appointment)  
David V. Bugg\*  
(graduate student from  
Cambridge Univ., England)  
Robert E. Chrien  
(postdoctoral appointment)  
Eugene L. Church  
(assigned from Frankford Arsenal)  
Victor W. Cohen  
Carl H. Collie\*  
(on leave from Oxford Univ., England)  
Rodney L. Cool  
Hans W. Courant\*  
(on leave from Yale Univ.)  
James W. Cronin  
Arthur C. Damask  
(assigned from Frankford Arsenal)  
Horace R. Danner\*  
(on leave from Pennsylvania State Univ.)  
Edward der Mateosian  
George J. Dienes  
Fahri Domanic  
(on leave from Univ. of Ankara, Turkey)

\*Terminated before June 30, 1958.

Alexander J. Elwyn  
(postdoctoral appointment)  
Albert R. Erwin, Jr.  
(graduate student from Harvard Univ.)  
Alonzo Fairbanks  
Gerald Feinberg  
(postdoctoral appointment)  
Ted B. Flanagan\*  
(assigned from Picatinny Arsenal)  
John D. Fox  
B. Chalmers Frazer  
Ananda M. Ghosh  
(on leave from Bose Research Inst., India)  
John B. Gibson  
Donald A. Gilbert\*  
G. Norris Glasoe  
Allen N. Goland  
(postdoctoral appointment)  
Gertrude S. Goldhaber  
Maurice Goldhaber  
Bernard P. Gregory  
(on leave from  
Ecole Polytechnique, France)  
Lee Grodzins  
Feza Gürsey  
(on leave from  
Science Univ. of Istanbul, Turkey)  
Rudolph Haas  
(on leave from Technische  
Hochschule, Munich, West Germany)  
Eastman N. Hatch\*  
(postdoctoral appointment)  
Robert D. Hatcher  
(on leave from NYU)  
John W. Hess  
(assigned from Picatinny Arsenal)  
John Hornbostel  
Donald J. Hughes  
Joseph Jach  
(assigned from Picatinny Arsenal)  
Maurice R. Jacob  
(postdoctoral appointment)  
John V. Kane  
Thomas A. Kaplan  
(on leave from Univ. of Michigan)  
Mortimer I. Kay  
(on leave from Pennsylvania State Univ.)  
David T. Keating  
Ottmar C. Kistner  
(graduate student from Columbia Univ.)  
Walter Kley  
(on leave from Albert Ludwigs  
Universität, Freiburg, West Germany)  
Joshua K. Kopp  
(graduate student from Harvard Univ.)  
Walter L. Kosiba\*  
Harry H. Landon\*  
Paul W. Levy  
Seymour J. Lindenbaum  
Robert I. Louttit

James E. Mapes  
(assigned from Picatinny Arsenal)  
Harvey Marshak  
James A. Martin, Jr.\*  
Santos Mayo\*  
(assigned from the AEC of Argentina)  
Kirk McVoy, Jr.  
(postdoctoral appointment)  
Toshio Mitsui  
(on leave from Pennsylvania State Univ.)  
Michael J. Moravcsik\*  
(postdoctoral appointment)  
Thomas W. Morris  
Robert Nathans  
(on leave from Univ. of Pennsylvania)  
Shimon Ofer  
(postdoctoral appointment)  
Harry Palevsky  
Antonio Paoletti  
(assigned from Comitato  
Nazionale Ricerche Nucleari, Italy)  
Simon Pasternak  
(assistant editor of The Physical Review)  
Eugene T. Patronis  
(postdoctoral appointment)  
Israel Pelah\*  
(on leave from  
Univ. of Amsterdam, The Netherlands)  
Oreste Piccioni  
Stanley J. Pickart\*  
(assigned from United States Army)  
Charles E. Porter, Jr.  
(on leave to  
Los Alamos Scientific Laboratory)  
Lew F. Porter  
(assigned from U.S. Steel Corp.)  
David C. Rahm  
Ralph R. Rau  
Kurt Reibel  
(graduate student from  
Univ. of Pennsylvania)  
H. Juergen Rose\*  
(postdoctoral appointment)  
David B. Rosenblatt  
(assigned from Frankford Arsenal)  
Brice M. Rustad  
(on leave from Columbia Univ.)  
George J. Safford  
(graduate student from Columbia Univ.)  
Vance L. Sailor  
Edward O. Salant  
Jack Sandweiss\*  
(on leave from Yale Univ.)  
Jack Schwartz  
(postdoctoral appointment)  
Robert B. Schwartz  
Arthur Z. Schwarzschild  
(postdoctoral appointment)  
Mary J. Scott\*  
(graduate student from  
Johns Hopkins Univ.)  
Ferdinand J. Shore, Jr.

Ralph P. Shutt  
Joseph E. Smith  
Lincoln G. Smith  
(on leave to Princeton Univ.)  
Hartland C. Snyder  
Andrew W. Sunyar  
Leon Tenzer\*  
(on Belgian-American  
Education Foundation Fellowship)  
Alan Thorndike  
George L. Trigg  
(on leave from Oregon State College)  
George H. Vineyard  
Joseph B. Vise  
(graduate student from Columbia Univ.)  
Tsunetaka Wajima  
(graduate student from Columbia Univ.)  
Ernest K. Warburton\*  
(postdoctoral appointment)  
Joseph Weneser  
Gian-Carlo Wick  
William L. Whittemore  
(assigned from General Dynamics Corp.)  
Robert L. Zimmerman  
Gus T. Zorn  
(on leave to Max Planck Inst.,  
Göttingen, West Germany)

### Reactor Division

Robert W. Powell, Head  
Sidney C. Abrahams  
(assigned from Bell Telephone Co.)  
John E. Binns  
Thomas H. Broome\*  
(assigned from The Martin Co.)  
John A. Dooley  
(assigned from  
Wright Air Development Center, USAF)  
Anthony N. Fasano  
(assigned from  
Wright Air Development Center, USAF)  
John J. Floyd  
H.K. Alan Kan  
(assigned from  
American Nuclear Science Corp.)  
Raymond H. Kelley  
(assigned from  
Wright Air Development Center, USAF)  
Gerald C. Kinne  
Issai Lefkowitz  
(assigned from Glenco Corp.)  
Walter Lones  
Leroy R. Lynam  
(assigned from  
Westinghouse Electric Corp.)  
Robert A. Meyers\*  
(assigned from  
Walter Kidde Nuclear Laboratory)  
Charles L. Osborne  
Howard Paitchel  
(assigned from Ford Instrument Co.)

\*Terminated before June 30, 1958.

Jack E. Phillips  
dePuyster G. Pitcher  
Edward Prince  
(assigned from Bell Telephone Co.)

\*Terminated before June 30, 1958.

Robert D. Schamberger  
(assigned from  
Wright Air Development Center, USAF)  
Ralph E. Segel  
(assigned from  
Wright Air Development Center, USAF)

Louis M. Slater  
(assigned from  
American Nuclear Science Corp.)  
Leon N. Zadoff\*  
(assigned from Ford Instrument Co.)