

# BROOKHAVEN NATIONAL LABORATORY



## *ANNUAL REPORT* *July 1, 1960*

Associated Universities, Inc.  
under contract with the  
United States  
Atomic Energy Commission

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A S S O C I A T E D U N I V E R S I T I E S , I N C .

## *Annual Report*



*July 1, 1960*

# BROOKHAVEN NATIONAL LABORATORY

Upton, Long Island, New York



Brookhaven National Laboratory is operated under a contract between the United States Atomic Energy Commission and Associated Universities, Inc. This, the eleventh in a series of unclassified Annual Reports, gives an account of the progress of the Laboratory during the period July 1, 1959 – June 30, 1960, 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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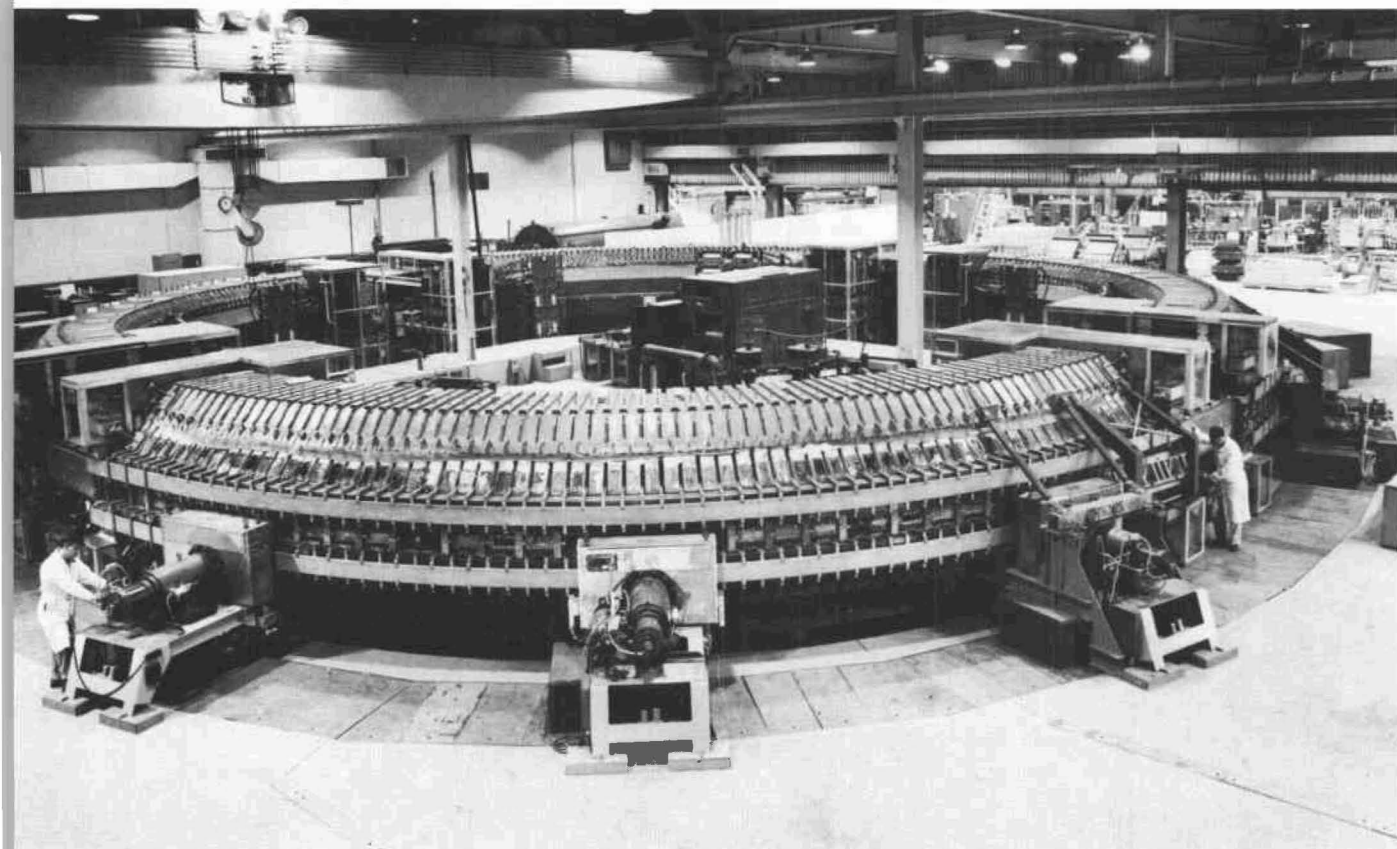
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The Cosmotron, before installation of the shielding. At the rear is a portion of the newly completed experimental area.

# Introduction

The program and activities of Brookhaven National Laboratory during fiscal 1960 are described in this annual report. The progress and trends of the research program are presented together 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 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 the report.

## PRESENT RESEARCH FACILITIES

Research at Brookhaven is centered on, although not confined to, the use of several large machines and other special facilities, some of which are described briefly below.

### Graphite Research Reactor

This reactor continues to support a major fraction of the research effort at Brookhaven. It has been in almost continuous operation since initial start-up in 1950. Its in-pile and beam facilities have been used in a variety of programs both by Brookhaven personnel and by visiting scientists and engineers from other institutions. Since 1957, a fuel reloading program has been under way, designed to replace the natural uranium fuel elements with new enriched fuel elements ( $>90\%$   $U^{235}$ ). The new burn-up cycle made it necessary during the past year to increase the number of loaded channels from 479 to 548. The maximum power of the reactor was increased from 17 Mw to 19 to 20 Mw, while the maximum thermal neutron flux remained at  $\approx 2 \times 10^{13}$  neutrons/cm<sup>2</sup>-sec. As a result of this reloading program, which may now be considered completed, the volume and pressure loss requirements for the cooling air were decreased sufficiently to make possible the replace-

ment of three of the five primary fans with fans designed to meet the new specific operating requirements. The savings in electrical power realized from operating with the new fans are expected to equal their cost within two years.

All the experimental facilities of the reactor, with the exception of some space on the top, have been fully utilized. Experimental space was made available to outside research organizations to the extent compatible with the requirements of the Laboratory's research program. Significant use was made of the reactor for some 16 outside projects; in addition, a considerable number of service irradiations were performed for outside organizations, and processed radioisotopes were also provided.

### Medical Research Reactor

The Medical Research Reactor (MRR), which first became critical on March 15, 1959, was constructed for the sole purpose of exploring the possible applications of nuclear reactors to the study of man and his diseases. Each salient feature of the reactor was designed in relation to its use for therapy and diagnosis or in the advancement of basic medical science. Since the initial start-up, continuous improvement has been made in the arrangements for the reflector and for the filtering and directing elements along the path of the neutron beam. The AEC has now granted permission to extend the maximum operating power level of the MRR from 3 to 5 Mw, provided that continuous operation at this new level does not exceed a period of 10 min. The limitation on excess reactivity was raised from 1 to 3%. Four brain tumor patients in far advanced stages of the disease were treated at the MRR by neutron capture therapy. One of these patients survived for 18 months, with minimum impairment and maximum effectiveness, following a single treatment. This is the maximum longevity attained thus far in cases of glioblastoma multiforme.

### Hot Laboratory

The Hot Laboratory, which is adjacent to the Graphite Research Reactor, contains extensive

facilities for the analysis, processing, and development of highly radioactive materials. It includes three hot cells in which chemical operations can be performed remotely while under observation by periscope. A larger hot cell of the cave type has been added recently for the physical examination of materials, especially metals, of high activity. Five semihot cells are used for work on less radioactive materials.

### **Critical Assembly Laboratory**

A critical assembly laboratory is available for research in reactor physics and for reactor development studies. It comprises two critical assembly areas, each with an operating console and assembly room. One of these rooms contains a small, water-cooled, graphite-reflected neutron source reactor for exponential studies. The laboratory also includes a pulsed Van de Graaff neutron source for diffusion experiments and reactivity measurements, an analogue computer for reactor kinetic studies, a subcritical assembly area, a dry-box room, and a necessary minimum of shop and office space. A thin-walled shed of the silo type has been added to the facilities; here measurements can be made of bucklings of bare homogeneous or semihomogeneous critical assemblies, with a minimum of neutron reflection from the ground and walls. An interior aluminum framework provides for the installation of critical experiments 20 ft from the floor. The first runs with a graphite moderator in this new facility are expected to take place late in 1960.

### **Cosmotron**

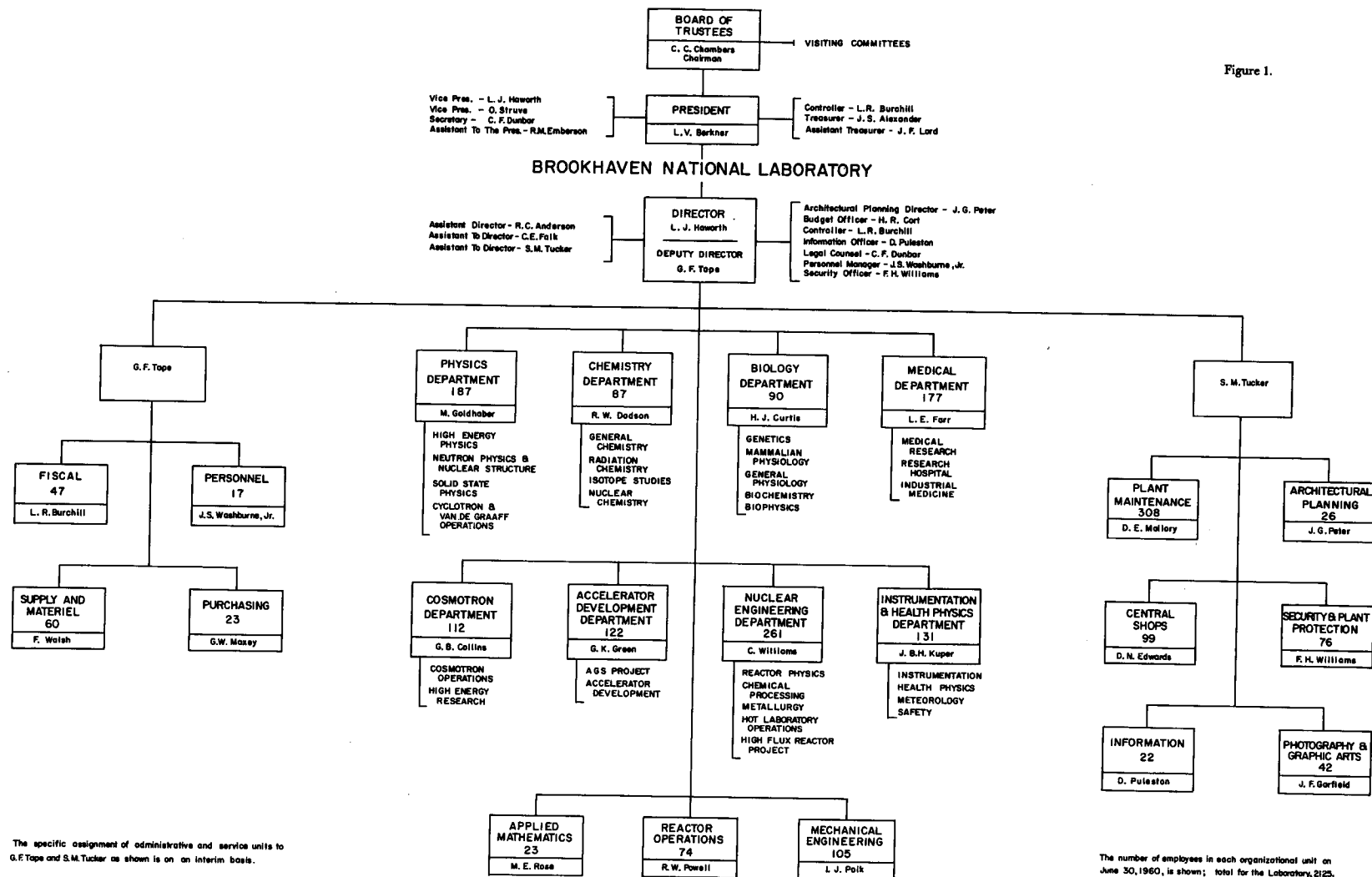
The past few years have seen marked changes in the character of the experiments performed at the 3-Bev proton synchrotron known as the Cosmotron. Interest is now centered on experiments designed to yield quantitative results of a specific nature, rather than on exploratory experiments of the type conducted several years ago. Quantitative experiments require intense, well-analyzed beams of high energy protons or pions, and these in turn call for elaborate arrays of electromagnets or other analyzing equipment, as well as massive shielding. These requirements have led to construction of the new experimental area and enlargement of power sources and other facilities, so that the accelerator itself is no longer predominant, but only part of a large Cosmotron complex.

Research operations were resumed at the Cosmotron in November 1959 after an extended shutdown for coil repairs and facility modifications. After four months of effective operation, the machine was again temporarily shut down because of the failure of some generator windings in the original power supply. By mid-1960 the Cosmotron was operating with reasonable reliability on a schedule of five 24-hr days per week, with  $\approx 80\%$  of this time available for experiments. Figure 3 shows an experimental setup in one of the external proton beams; from it an idea can be gained of the complexity now characteristic of experiments and of the resources available. In addition to the hydrogen bubble chamber shown in the background, this experiment involved eight electromagnets and a beam separator, all concealed by the concrete and steel shielding blocks. Similar setups are possible along any one of the three external proton beams, and thus an experiment can be assembled in one beam while a second experiment is being carried out in one of the other locations.

The Cosmotron is producing reliable internal beams of  $\approx 10^{11}$  protons per pulse. It is hoped that a circulating beam of  $10^{12}$  protons per pulse can eventually be obtained after several known limitations to the beam intensity have been corrected. The first difficulty stems from distortions now existing in the Cosmotron's magnetic field, caused by iron devices, such as the deflection magnets for the external beams, and the magnetic shield for the ferrite used to accelerate the protons, all added during the past few years. These distortions are believed to have accentuated unwanted resonances which prevent the use of the whole vacuum chamber during acceleration. They occur principally in the straight sections between the magnet quadrants and can be corrected by magnetically shielding each straight section with a 2 to 3-in.-thick iron box. The second limitation on the beam intensity appears to result from space charge formed by the circulating protons soon after their injection into the vacuum chamber. Studies of this interesting and important phenomenon are continuing.

The supplementary services at the Cosmotron facility include both technical assistance and the availability of electromagnets and liquid hydrogen. Experimental groups also can arrange to use more specialized equipment, which is maintained and operated by Laboratory personnel. Now

# ASSOCIATED UNIVERSITIES, INC.



## ORGANIZATION CHART

JUNE 30, 1960



**Table 1**  
**Organizational Expenditures - Fiscal 1958, 1959, 1960**  
(Includes Operating, Services to Fixed Assets, and Work for Others.  
Direct Costs of AGS and Other Fixed Assets and Additions to Inventory Are Not Included;  
See Tables 3 & 4)

		Salaries, Wages, Insurance								% of Total	Man-Years		
		Staff	Consultants & Temporary Employees	Travel	Material & Supplies	Subcontracts & Special Procurements	Power	Miscellaneous (Net)	Total Organizational Costs		Scientific (Incl. Guests)	Others	Total
Physics & Chemistry Research	1960	3,867,825	202,370	129,020	1,088,434	130,681	130,386	—	5,548,716	26.5	232.5	284.0	516.5
	1959	3,061,815	136,965	121,780	948,083	327,064	24,527	—	4,620,234	24.8	209.0	202.0	411.0
	1958	2,533,881	127,662	94,859	683,537	298,681	31,682	—	3,770,302	21.3	210.0	171.5	381.5
Biology, Medicine & Biophysics Research	1960	2,002,118	77,678	79,408	580,596	2,562	—	(5,158)	2,737,204	13.1	96.5	196.5	293.0
	1959	1,799,455	60,616	86,270	464,580	21,324	—	(10,920)	2,421,325	13.0	97.5	180.0	277.5
	1958	1,457,015	60,152	72,079	330,976	36,809	—	(8,001)	1,949,030	11.0	85.0	159.5	244.5
Nuclear Engineering Research	1960	1,663,191	44,176	46,563	436,454	148,173	(179)	—	2,338,378	11.2	74.0	124.0	198.0
	1959	1,655,173	39,546	66,091	491,674	536,224	10,154	—	2,798,862	15.0	79.5	151.0	230.5
	1958	1,425,945	32,136	51,368	426,081	1,662,173	—	—	3,597,703	20.3	83.5	190.0	273.5
Isotope Development	1960	96,401	3,047	7,947	66,538	13,396	—	—	187,329	0.9	4.0	7.0	11.0
	1959	27,830	—	3,339	20,631	6,314	—	—	58,114	0.3	1.5	2.0	3.5
	1958	—	—	73	189	22,512	—	—	22,774	0.2	—	—	—
Training & Education	1960	27,180	411	32,736	44,486	4,277	—	—	109,090	0.5	2.0	4.0	6.0
	1959	11,974	3,924	3,703	4,545	—	—	—	24,146	0.1	5.5	—	5.5
	1958	19,324	8,220	9,025	2,028	36,786	—	—	75,383	0.4	8.5	0.5	9.0
Radiation Protection	1960	365,589	2,886	2,838	88,498	—	—	(1,985)	457,826	2.2	6.5	44.0	50.5
	1959	312,931	1,395	1,876	51,063	—	—	(3,675)	363,590	1.9	8.5	38.0	46.5
	1958	291,109	1,284	1,858	43,421	—	—	(1,573)	336,099	1.9	11.0	36.5	47.5
Supporting Scientific & Technical Services	1960	2,683,242	15,364	25,108	460,831	—	390,407	(191,770)	3,383,182	16.1	24.5	275.5	300.0
	1959	2,374,179	12,742	16,680	355,729	3,360	400,365	(278,709)	2,884,346	15.5	26.5	304.0	330.5
	1958	2,066,535	16,890	18,975	346,487	69,299	420,449	(256,551)	2,682,084	15.1	29.5	287.0	316.5
Security & Plant Protection	1960	563,172	754	795	13,371	—	—	2,894	580,986	2.8	—	77.5	77.5
	1959	528,953	698	609	7,240	—	—	2,407	539,907	2.9	—	80.0	80.0
	1958	503,223	133	896	12,681	—	—	2,552	519,485	2.9	—	81.0	81.0
Miscellaneous (including Lighting, T & T, Heating Fuels, Special Maintenance, etc.)	1960	—	—	2,416	175,618	—	347,818	728,851	1,254,703	6.0	—	—	—
	1959	—	—	653	158,757	—	293,720	707,613	1,160,743	6.2	—	—	—
	1958	—	—	495	118,650	—	289,875	956,898	1,365,918	7.7	—	—	—
General and Administrative	1960	3,424,033	19,715	92,011	559,725	(9,211)	—	(65,642)	4,020,631	19.2	5.0	546.5	551.5
	1959	3,144,536	23,936	80,034	369,155	2,554	—	(139,346)	3,480,869	18.7	7.0	479.0	486.0
	1958	2,747,959	36,092	62,110	369,673	—	—	(109,493)	3,106,341	17.6	—	453.5	453.5
Laboratory Total	1960	14,692,751	366,401	418,842	3,514,551	289,878	868,432	467,190	20,618,045	98.5	445.0	1,559.0	2,004.0
	1959	12,916,846	279,822	381,035	2,871,457	896,840	728,766	277,370	18,352,136	98.4	435.0	1,436.0	1,871.0
	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	1,319.5	1,747.0
AUI Administration	1960	—	—	—	—	—	—	178,000	178,000	0.8	—	—	—
	1959	—	—	—	—	—	—	178,100	178,100	0.9	—	—	—
	1958	—	—	—	—	—	—	158,000	158,000	0.9	—	—	—
Total AUI and BNL	1960	14,692,751	366,401	418,842	3,514,551	289,878	868,432	645,190	20,796,045	99.3	445.0	1,559.0	2,004.0
	1959	12,916,846	279,822	381,035	2,871,457	896,840	728,766	455,470	18,530,236	99.3	435.0	1,436.0	1,871.0
	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	1,319.5	1,747.0
Work for Others, Direct Costs Only	1960	64,051	2,289	4,841	60,127	7,158	—	—	138,466	0.7	6.0	6.5	12.5
	1959	49,964	2,031	1,815	68,395	549	—	8,314	131,068	0.7	4.5	4.5	9.0
	1958	53,923	72	4,333	65,433	420	—	—	124,181	0.7	9.0	8.0	17.0
Grand Total	1960	14,756,802	368,690	423,683	3,574,678	297,036	868,432	645,190	20,934,511*	100.0	451.0	1,565.5	2,016.5
	1959	12,966,810	281,853	382,850	2,939,852	897,389	728,766	463,784	18,661,304**	100.0	439.5	1,440.5	1,880.0
	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	1,327.5	1,764.0

\*\$553,081 of this total was distributed to Fixed Assets and as services to Work for Others and Inventory.

\*\*\$420,606 of this total was distributed to Fixed Assets and as services to Work for Others and Inventory.

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

available are 20, 14, and 12-in. hydrogen bubble chambers, and soon 30-in. propane and 30-in. hydrogen bubble chambers will be added. A beam separator that separates particles of the same charge and momentum but of different mass has recently been placed in operation. Figure 5 shows this device, which operates on the same principle as a mass spectrometer by using crossed magnetic and electric fields.

#### 60-in. Cyclotron

There have been no major changes in this cyclotron, which accelerates helium nuclei to 42 Mev, deuterons to 21 Mev, and protons to 10.5 Mev. It has been in regular operation for a wide variety of activities in chemistry, physics, medicine, and biology, and for certain technical applications. More than 40 experimenters from 5 of the Laboratory's departments and 16 outside institutions have used the machine. The outside institutions accounted for 37% of the operating time.

Minor changes and additions to the cyclotron are required from time to time to meet the demands of the experimental program. A beam measuring and monitoring system is currently under development as a prelude to energy stabilization; more extensive modifications are being considered. The erosion of water-cooling lines,

particularly in the dees, has continued to be a problem. New dee lines have been designed and are being fabricated.

#### 18-in. Cyclotron

The 18-in. cyclotron, formerly housed in the test shack in back of the Cosmotron, is now being used for research in its new location in the recently completed extension to the Cyclotron-Van de Graaff Building. This machine accelerates protons up to 3-Mev energies, and several recent improvements have made it possible to focus a 100- $\mu$ a beam of 3-Mev protons on a spot  $< \frac{3}{8}$  in. in diameter at the target position 30 ft from the machine. Voltage stabilization for several of the cyclotron components, particularly the high voltage supply for the rf oscillator and the power supplies for the focusing magnets, has eliminated the perturbations previously experienced as a result of line voltage fluctuations. It is planned to continue the preliminary work on studies of elastic and inelastic scattering of polarized neutrons in the energy range from several hundred kev to  $\approx 2$  Mev.

#### Electrostatic Generator

This accelerator provides proton beam currents up to 100  $\mu$ a. With the installation of the new accelerator tube, the machine has operated re-

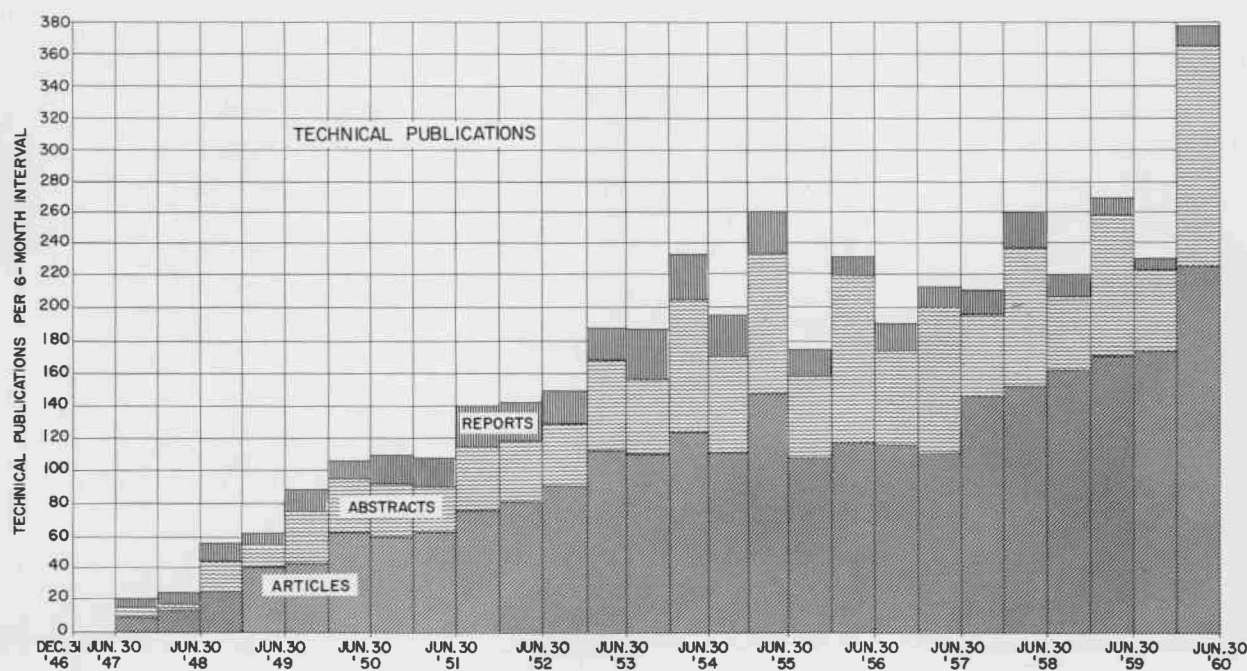


Figure 2.

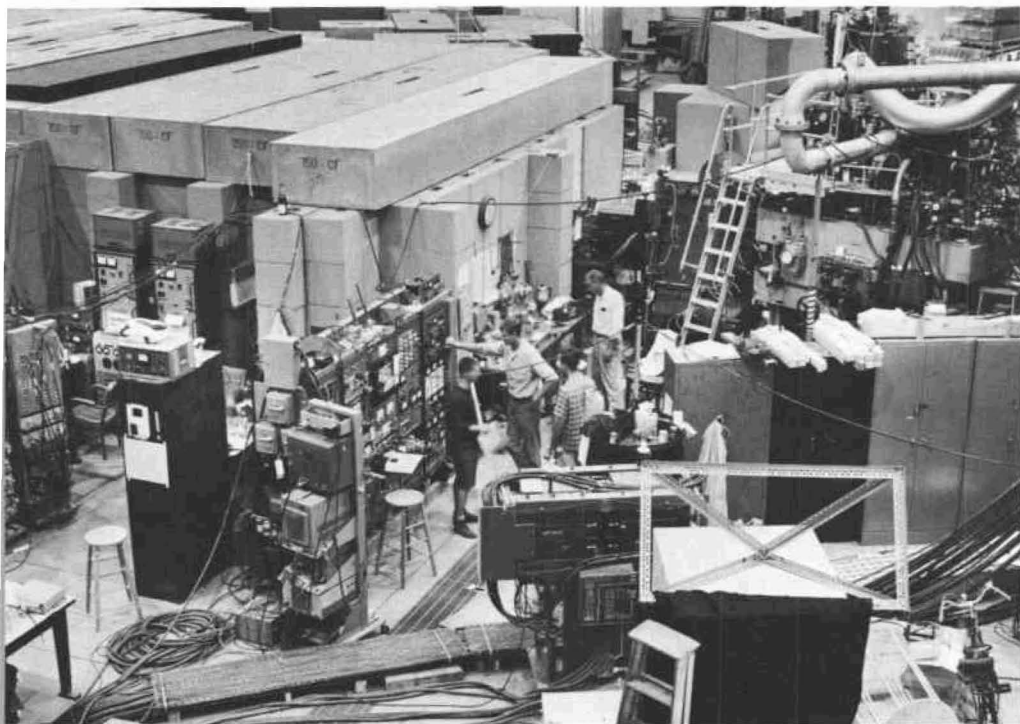
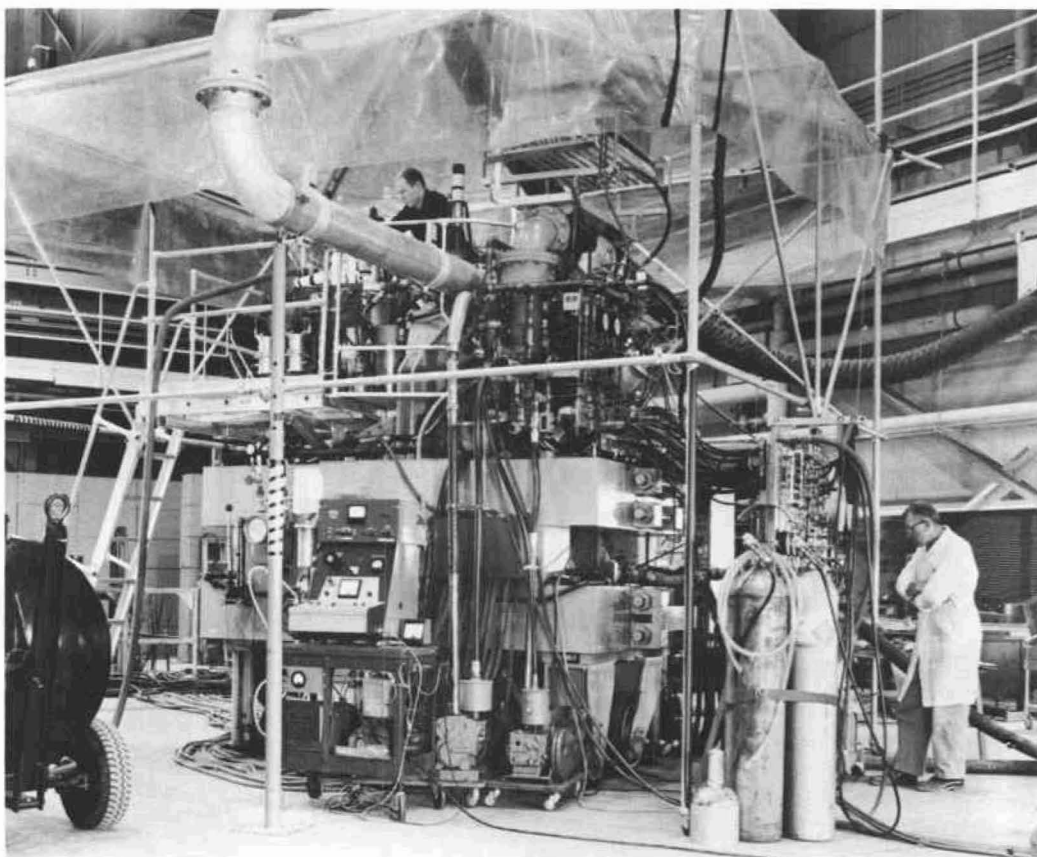


Figure 3. Experimental setup at Beam 2 in the Cosmotron.

Figure 4. The Brookhaven 20-in. liquid hydrogen bubble chamber, installed in the new experimental area at the Cosmotron.



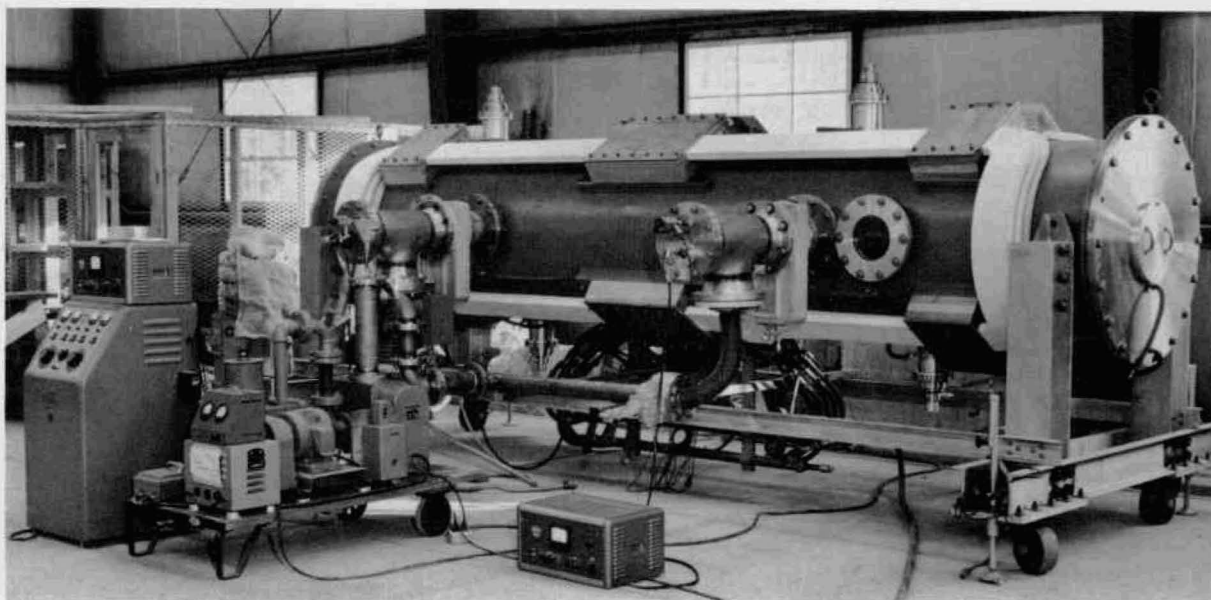


Figure 5. Cosmotron beam separator No. 1.

liably up to an energy of 3 Mev. The experience obtained from this operation has indicated the desirability of definite design modifications to raise and maintain the upper limit on the voltage. The modifications are now being designed and will be incorporated when it is feasible to interrupt machine operation.

This Van de Graaff machine was used for research on 194 of a possible 255 days during the year. The installation of the new tube required a major shut-down of 46 days; if this is excluded, the machine continued to have an operational efficiency of  $\approx 90\%$ . Research in physics accounts for  $\approx 90\%$  of the machine's time, the remaining 10% being used for continuing research by the Medical Department on the irradiation of mice with fast neutrons.

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 Department. In general, this machine is used in studies of the chemical effects produced by radiations.

#### GENERAL CONSTRUCTION PROGRESS

The complex of structures originally designed to house the Alternating Gradient Synchrotron (AGS) and its ancillary equipment has been completed. An addition to one wing of the Service

Building is also ready for occupancy as office space. To meet the growing requirements of the research program being planned for this accelerator, an addition to the existing experimental area, consisting of a 60,000 square-foot L-shaped extension of the Target Building, is now being designed. In addition, preliminary designs have been started for a building for the 80-in. bubble chamber facility, which will be located on the outer side of the AGS tunnel, north of the Target Building. A 9600 square-foot addition to the laboratory area of the Service Building will also be constructed.

The new experimental area at the Cosmotron was completed. This  $100 \times 180$ -ft room has a height of 60 ft and is serviced by a 40-ton overhead crane for the installation and moving of bubble chambers, magnets, shielding blocks, and other heavy equipment. The new area is supplied with the large sources of electrical power and cooling water required by the numerous electromagnets essential to the experimental external beam setups. The Cosmotron itself is now covered with a massive shield of heavy concrete 6 to 9 ft thick that weighs 8000 tons. This increased shielding is necessitated by the expected circulating beam intensity of  $10^{12}$  protons per pulse.

As part of a long-range site and building program known as BNL Master Plan - 1958, construction was begun on two buildings to house the major activities of the Nuclear Engineering and



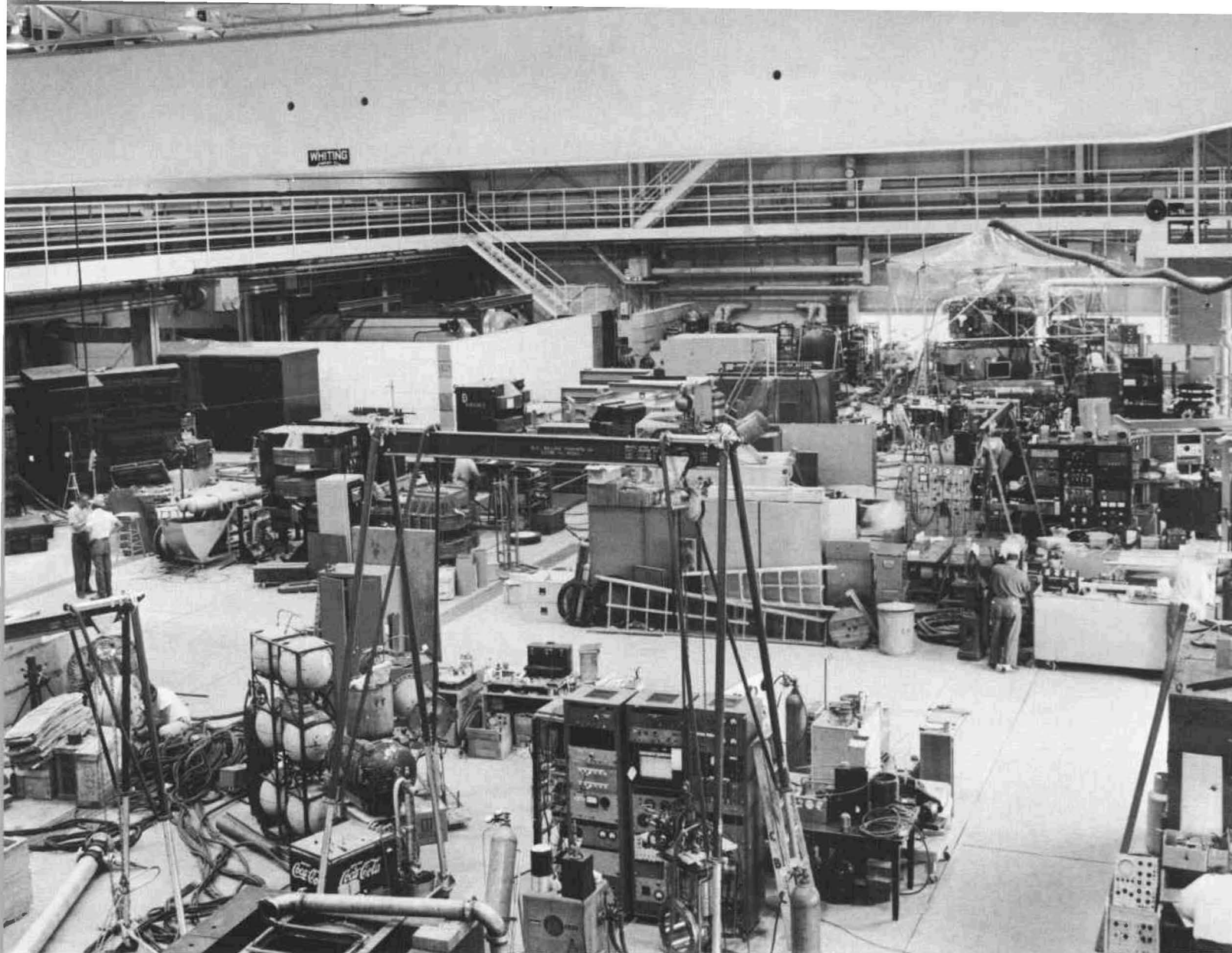


Figure 6. New experimental area at the Cosmotron, where bubble chambers and ancillary equipment are in process of installation. At the right rear is the Brookhaven 20-in. liquid hydrogen bubble chamber.

Physics Departments. To date, various sections of these two departments have been scattered in a number of temporary structures, many of them converted World War II barracks-type wooden buildings, inefficient in operation and increasingly costly to maintain. Construction of the first phase of the Nuclear Engineering Building was begun in mid-1959; this phase, with an area of  $\approx 38,000$  square feet, will accommodate most of the Department's activities in the fields of chemistry and chemical engineering. Occupancy is expected early in 1961. An artist's concept of this building is shown in Figure 7. In addition, funds have been appropriated and design has been started on a

small volatility studies laboratory and on additions to the critical assembly area and the Metallurgy Building.

In March 1960, ground was broken for the new Physics Building, which will provide an area of  $\approx 104,000$  square feet of laboratory and office space for the activities of the Physics Department, now housed in ten temporary buildings. The new building will include a two-story laboratory section containing 27 general laboratories, together with special laboratories for the programs in nuclear moments and solid state physics, for scanning and data analysis work connected with the emulsion and bubble chamber programs in high



Figure 7. Artist's concept of the first phase of the Nuclear Engineering Building. This phase, now under construction, will provide a gross area of 38,000 square feet.



Figure 8. Artist's concept of the Physics Building, on which construction was begun in March 1960. When completed in about two years, the building will provide an area of  $\approx 104,000$  square feet for laboratories, offices, conference rooms, and a library.

energy physics, a computer room, and a drafting room. Some of the laboratories will be equipped for work with radioactive materials and will be shielded from areas requiring a low radiation background. Construction should be completed in about two years. An artist's concept of the building is shown in Figure 8.

## **MAJOR RESEARCH FACILITIES UNDER DESIGN OR CONSTRUCTION**

### **Alternating Gradient Synchrotron**

All the major components of this major research facility, on which work was begun in 1954, have been completed and assembled, and preliminary tests of the 30-Bev AGS are in process. The 50-Mev linear accelerator injector (linac) has undergone extensive testing as an operating unit, and on May 17, 1960, a 50-Mev proton beam was injected into the synchrotron and successfully completed one turn around the magnet ring. Nine days later a beam was successfully spiraled around the ring  $\approx 100$  times. Detailed plans have been made for the first system testing of the entire synchrotron. Before the AGS can be used for experimental work, many measurements must be made of the machine parameters to explore and understand fully its behavior.

Precise surveys of the main magnet foundations indicated maintenance of the desired stability, with no changes  $>0.010$  in. per year except where the loads were altered. The data from these surveys were used to realign the magnets radially and vertically prior to the final assembly of the vacuum chamber. Because of the expected sensitivity to future changes in loading, check surveys will be made before and after future construction and the installation of experimental equipment.\*

### **High Flux Beam Research Reactor**

All the essential features and most of the minor components of the High Flux Beam Research Reactor (HFBR) have now been determined. Approximately 700 critical experiments were performed in reaching the final design details. In most of these experiments the reactor core or structural components, or both, had been changed, to determine the effect of these changes on the re-

activity. Some of the experiments gave information on such features of neutron performance as power and flux distributions and neutron lifetime. A rough mock-up containing the core and structural components has been assembled and tested. The next step is the assembly of a precise and final mock-up, which will undergo a complete set of measurements on excess reactivity, poison reactivity effects, flux distribution, and other parameters. From these data it will be possible to determine with precision the neutron performance of the reactor.

The reactor core is cooled, moderated, and reflected with heavy water. It consists of 28 MTR-type fuel elements housed in a shroud in the lower, spherical portion of the reactor vessel. Cool  $D_2O$  is pumped into the upper, cylindrical part of the vessel, flows downward inside the shroud through the fuel elements, and is discharged into the bottom of the vessel. It then flows upward outside the core and leaves from an outlet pipe at the upper part of the vessel, to be cooled by heat exchange with water from cooling towers. Outside the core vessel is a water-cooled thermal shield of steel plates and lead-cadmium alloy. This secondary vessel provides emergency containment to keep the core covered with  $D_2O$  in the event of a leak in the primary vessel. Sixteen control rods, actuated by drive mechanisms on the upper portion of the reactor vessel, are located in the reflector.

The HFBR will be housed in a three-story, circular, domed, gas-tight building. The bottom floor will house the operating machinery for the reactor; the second or ground-level floor will be reserved for beam experiments and laboratories; and the top floor will accommodate the control room, irradiation experiments, and fuel handling facilities.

This reactor, which is expected to deliver a peak thermal flux of  $\approx 5$  to  $10 \times 10^{14}$  neutrons/cm<sup>2</sup>-sec and epithermal fluxes of  $\approx 1$  to  $2 \times 10^{15}$  neutrons/cm<sup>2</sup>-sec, will be used primarily as a source of high intensity external thermal and epithermal neutron beams for studies in the physical sciences.

### **Merlin - A Digital Computer**

The construction phase of this large and versatile high-speed computer of advanced design was completed during the year, and the machine became available for preliminary testing and subsequently for the development of the programming system. Work was begun on the coding of

\*Since the end of the fiscal year, the AGS has successfully accelerated protons to full design energies. This was first done on July 29, 1960, when energies above 30 Bev were obtained.

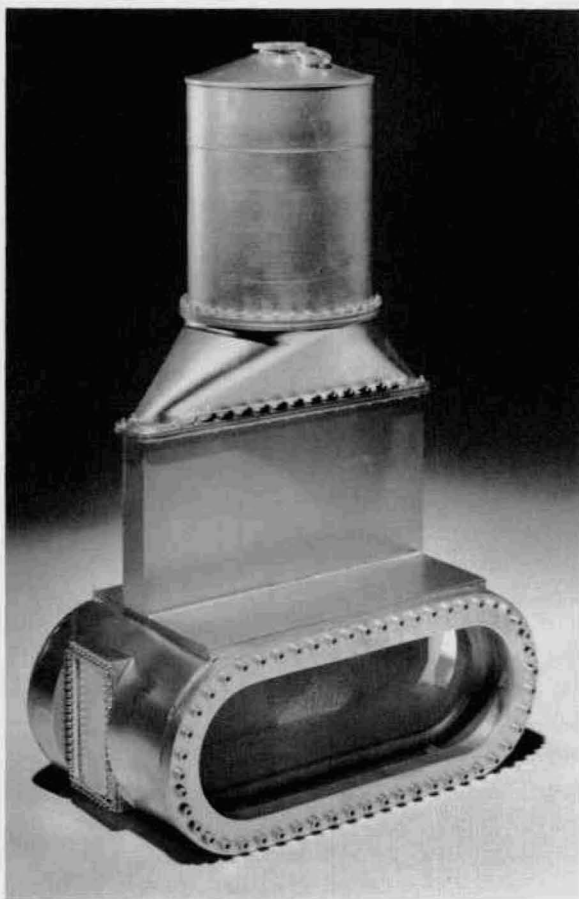


Figure 9. Model of the 80-in. bubble chamber body. The glass window in front is for illumination and photography, and at each side is a thin metal beam window. The 36-in.-diameter cylinder on top contains the piston used for expanding the chamber. The entire assembly, weighing  $\approx 10$  tons, will be made of stainless steel.

specific problems, such as the preparation of a multigroup reactor code and of a Monte Carlo code for the study of high energy nuclear reactions. It is expected that Merlin will be available for full-scale computational services to the Laboratory's research program late in 1961.

#### 80-in. Bubble Chamber

A large, liquid hydrogen bubble chamber for use with the AGS is now being designed and built (see Figures 9 and 10). The stainless steel chamber,  $80 \times 25$  in. and 28 in. deep, will have a useful hydrogen volume of 900 liters and will be expanded by a 36-in.-diameter piston which can be recycled once per sec. A  $6\frac{1}{2}$ -in.-thick half-tempered glass plate will provide a window for illumination and

photography. Calculations and model experiments indicate that the magnet, which will contain 30 tons of water-cooled copper windings and 280 tons of magnetic return iron, should have a field of 18,000 gauss when powered with 4 Mw.

The chamber will slide on rails in a special building located at the outside of the AGS magnet tunnel. This building will house all the facilities needed to operate the chamber, including a 40-ton crane in the  $70 \times 60$ -ft main hall; a compressor room for the operation of hydrogen, nitrogen, and Freon refrigerators and the expansion system; electronic control rooms; and a machine shop. Beams will be guided into the chamber through a new opening in the AGS tunnel. Contracts for the major components of the bubble chamber facility

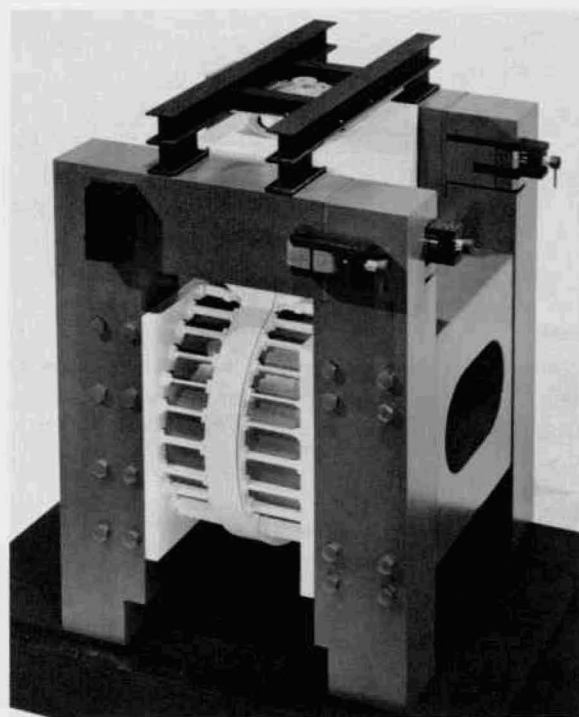


Figure 10. Model of the magnet for the 80-in. bubble chamber, showing the 280-ton steel yoke and the 30-ton copper coils. The large opening at the right side provides for photography and illumination. Between the coils and near the top of the magnet are seen portions of the vacuum chamber, which provides thermal insulation around the chamber containing the liquid hydrogen at a temperature of  $\approx 27^\circ \text{K}$ . The chamber hangs from the I-beam bridge at the top. Also shown are some of the many jacks that will hold together the two sides of the assembly; separation of the sides will be necessary when work on the chamber is being performed.



will be let in the near future, and operation is expected to start in 1962.

## RESEARCH ACTIVITIES

The Brookhaven research program covers a wide range of subjects in the physical and life 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 that constitute them, and the forces involved in their structure.
2. Studies of the physical, chemical, and biological effects of nuclear radiation.
3. The use of nuclear tools, such as neutrons, charged particles, gamma-rays, and isotopic tracers, in all branches of scientific research.
4. Research and development, not necessarily itself of a nuclear nature, directed toward solving the problems of nuclear energy development.
5. The development of specific devices for use as research tools or in practical applications of atomic energy.

Given below are brief summaries of typical activities during the past year in these categories. More complete and detailed accounts of various aspects of the research program may be found in the sections of this report that follow the Introduction.

### 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 at 3-Bev energy from the Cosmotron to neutrons of  $<1$ -ev energy from the Graphite Research Reactor. The studies of these events are carried out not only by physicists but also by nuclear chemists. The work at high energy has attracted the interest and enlisted the active collaboration of scientists from a number of universities and research institutions in this country and abroad. This work has been centered on the Cosmotron; now that this accelerator is again functioning effectively and its experimental area has been expanded considerably, quantitative experiments with intense, well-analyzed beams of high

energy protons and pions are under way. Although photographic emulsion stacks and arrays of scintillation and Cerenkov counters are used, the most frequent practice is to use a variety of hydrogen and propane bubble chambers, some of which are still being installed in the new experimental area, to record the resulting events.

One group at the Cosmotron, working in the field of the so-called strong interactions that occur between nucleons and  $\pi$ -mesons, is studying the  $\pi^-p$  processes. In one of these reactions,  $\pi^- + p \rightarrow \pi^- + \pi^0 + p$ , support is found for the existence of a strong  $\pi$ - $\pi$  attraction, which holds the  $\pi^0$  and  $\pi^-$  together for a short time. A model that is acceptable so far presents the incoming  $\pi^-$  as striking a  $\pi^0$  existing in a virtual state near the proton and the two mesons then moving away together, leaving the proton relatively undisturbed. This investigation is being continued in further experiments.

In studies of strange particles, experiments to produce hyperons and  $K$ -mesons are in progress. About 3000 strange particle decays have been observed in the 14-in. hydrogen bubble chamber by using a pulsed beam containing  $\approx 3 K_2^0$ -mesons per Cosmotron pulse. So far, four  $\Xi^0$  hyperons have been found, which confirms the existence of this particle and amplifies the data obtained from the single event observed at Berkeley.

The program planned for the Cosmotron includes several bubble chamber experiments to study the process of associated production near threshold by  $\pi^+$  and  $\pi^-$  mesons. To obtain further information on  $\pi$ -nucleon scattering at the 1-Bev level, an extensive series of bubble chamber runs is also scheduled. Among future experiments with counters are the measurement of the magnetic moment of the  $\Lambda^0$ -hyperon, the determination of production cross sections for  $K$ -mesons and  $\pi$ -mesons from  $p$ - $p$  interactions, and studies of the production of  $K_2^0$ -mesons from  $\pi$ - $p$  interactions as a function of incident energy and angle.

The Cosmotron has also served as an important research tool for the nuclear chemists in studying nuclear reactions at Bev energy levels. The interactions of high energy protons with silver and bromine nuclei in emulsions reveal that with 1.0-Bev protons  $\approx 3\%$  of the interactions lead to fission, but that at 2.0 and 3.0 Bev this proportion increases to  $\approx 7$  and 11%, respectively. By measuring the angular distributions of the alpha and other light particles with respect to the fission fragments in these events, conclusions could be

reached about the time sequence of fission and particle emission which indicate that the latter precedes fission in most cases. Other studies are centered on accounting for the yield of light nuclides such as  $\text{He}^6$ ,  $\text{Be}^7$ , and  $\text{Li}^8$  by an evaporation mechanism in which these, along with neutrons, protons, and alpha-particles, are boiled out of highly excited nuclei resulting from the interaction of high energy protons with heavy target atoms. In more general studies on the expected behavior of recoiling nuclei in high energy reactions, the momentum imparted to such nuclei in the cascade process has been calculated by Monte Carlo methods for 0.46 to 1.84-Bev protons incident on  $\text{Ru}^{100}$ ,  $\text{Bi}^{209}$ , and  $\text{U}^{238}$ . Uranium fission by protons in the Bev range has also been under study. High energy pions from a high intensity beam from the Cosmotron were made available for the bombardment of mercury targets, and a number of reaction products were isolated. Radiochemical studies of these pion-induced reactions will be continued, in the expectation that the cross section pattern will be found to differ in some significant respects from that found in proton interactions.

With the availability of the AGS for experimental work next year, the Laboratory's high energy program will be greatly expanded. An area of high energy research heretofore accessible only on a limited scale through cosmic rays will be opened up with the 30-Bev protons from this machine. The AGS is expected to be an abundant source of antiparticles and also, through the decay of high energy  $\pi^0$ -mesons, of 1 to 25-Bev gamma-rays and electrons.

Research on the fundamental structure of atomic nuclei may be divided into two broad categories: studies of the characteristics of unstable nuclei, and studies of the characteristics of the instantaneous products of nuclear reactions. In the first category, information is sought on the ways in which an unstable nucleus is produced and then reverts to a stable nucleus, through the process of radioactive decay. Characteristics such as spin, parity, and lifetime are analyzed to determine the systematics of the decay process. In the second category, data are obtained on the characteristics of energy levels of nuclei by studying radioactive decay, or, in cases involving very short lifetimes, through study of nuclear reactions. The latest developments in electronic circuitry have made it possible to observe lifetimes as short as  $10^{-11}$  sec with high precision.

The recently discovered recoil-free emission and resonant absorption of gamma-rays when nuclei are tightly bound in a crystal lattice, known as the Mössbauer effect, has opened a new field of experimentation in the study of chemical binding effects and internal magnetic fields in materials. For example, the 14.4-kev nuclear gamma-ray of  $\text{Fe}^{57}$  has been used to determine the quadrupole coupling for the  $\frac{3}{2}$  — excited state of  $\text{Fe}^{57}$  bound in  $\text{Fe}_2\text{O}_3$ , and to measure an energy shift of this gamma-ray which can be attributed to binding effects.

A lower limit for the "lifetime" of the electron against decay has been established. When an electron disappears from the *K*-shell of an atom such as iodine, the process can be detected by the subsequent radiation of *K* x-rays when the vacancy is filled by an electron from an outer shell. The amount of *K* x-rays in iodine present in a large NaI(Tl) crystal was used to set a lower limit on any such process.

The charged particles available from the Van de Graaff accelerator were used for a number of problems relating to the structure of light nuclei. One of these problems concerned the energy levels in  $\text{C}^{12}$ , particularly the 7.66-Mev second excited state, which is thought to be involved in helium burning in red giant stars. There had been no experimental proof that carbon could be formed from helium, but after preliminary experiments here indicated that the ground-state transition might be detected in the  $\text{Be}^9(\alpha, n)\text{C}^{12}$  reaction, the intermediate-image pair spectrometer was moved to Oak Ridge National Laboratory in order to carry out the measurements with the high energy alpha-particles available from the ORNL Van de Graaff.

In a continuing program of research in neutron physics, the BNL Graphite Research Reactor has been used to study the interactions between cold neutrons and nuclei in liquids and solids. The neutrons, filtered through beryllium and beamed through the Brookhaven slow chopper, are scattered from liquid samples; thus, data are elucidated on the diffusive motions of liquid molecules at various temperatures. Use of the BNL fast chopper and time-of-flight analyzer has been mainly devoted to two areas of interest in neutron spectroscopy. The first of these is the remeasurement of total radiation widths that have been reported to fluctuate widely from level to level in the same nuclide, notably in the cases of antimony

and mercury. The second area of interest is a continuing study of the total cross section of nuclides with atomic weights in the range 100 to 120, especially in the cases of rhodium, tellurium, and ruthenium. In addition, the neutron widths and energies of individual levels have been obtained for these nuclides in the energy region from  $\approx 10$  to 400 eV above the neutron binding energy.

The accumulation of new data on neutron cross sections and the refinement of old measurements have made the publication of supplementary data to BNL 325, *Neutron Cross Sections*, a continuing project. The bulk of new data is in the area of capture cross sections.

In the untimely death from a heart attack of Donald J. Hughes on April 12, 1960, the Laboratory and the world of nuclear physics lost an outstanding scientist. Since joining the Brookhaven staff in 1949, Dr. Hughes had organized the group that collects and publishes all available information on the interactions of neutrons with matter. This compilation (BNL 325) has become a standard, world-wide source for scientists and engineers. Dr. Hughes played a unique role in the development of the world's current knowledge of the neutron and was one of the main U.S. scientific ambassadors responsible for the current fruitful international relationships in the fields of neutron physics and nuclear science. It is a testimonial to his abilities that the project Dr. Hughes established at Brookhaven is to be continued.

### Effects of Nuclear Radiation

This category includes studies of the physical, chemical, and biological effects of nuclear radiation. Results frequently reveal new information concerning the characteristics of the substances irradiated, or throw new light on the internal processes of living organisms and the changes they undergo in their life and reproductive cycles.

In the field of solid physics, most of the recent theoretical work has been in the radiation-induced production of defects in crystals. Improved computational procedures have made it possible to present a picture of the time sequences of damage events and processes. In the study of such events at low energy, this technique has been an important tool in revealing the intricate atomic motions associated with the interaction of radiation with matter, and it is now planned to adopt it for higher energy events. In the annealing of radiation-

induced or quenched-in defects, recent theoretical work has shown that impurities, even when present in very low concentrations, can seriously influence the annealing kinetics. The general equations for the annealing of vacancies in metals containing impurities to which the vacancies can be attached have been solved by analogue computer for a wide variety of parameters. It is shown that impurity contents as low as  $10^{-5}$  can markedly affect the results; furthermore, the calculations indicate that careful annealing experiments on impure metals prepared by controlled doping can be used to measure the binding energies of vacancies to impurities.

The major experimental research in solid state physics has been in studies of radiation effects and other departures from perfect periodicity, by many diverse techniques. Some of the investigations in this area are the search for experimental evidence of the production of thermal spikes during irradiation in other-than-fissionable materials; the study of radiation-enhanced diffusion in  $\alpha$ -brass, in cooperation with Los Alamos Scientific Laboratory, with a nuclear rocket engine used as a source of fast neutrons; the determination of density changes in diamonds irradiated at high neutron flux in the Materials Testing Reactor; optical measurements on alkali halides and other insulators, to determine the number and character of radiation-induced defects; the application of ultrasonic techniques in the study of radiation-induced changes in solids; and the effect of radiation on the chemical activity of various solids.

The chemical reactions in irradiated aqueous solutions are produced by short-lived intermediates (free radicals) which result from the excitation and decomposition of water. These reactions depend upon the number and nature of the intermediates, their spatial distribution, and the reactions they undergo with dissolved substances. In studying the chemistry of the reactive radicals, it is desirable to determine their absolute reaction rates and lifetimes. Research in this field is proceeding along three lines: (1) determination of reaction mechanisms in a variety of solutions, including the intricate reactions occurring among the decomposition products of pure water itself; (2) special experiments using pulsed radiation or flow systems designed to determine the lifetimes and hence the absolute reaction rates of the various radicals; and (3) experiments on the scavenging of free radicals at relatively high solute concentrations, in which

the details of the nature and initial distribution of the radicals are a predominant factor.

In studies on the radiolysis of organic compounds, the effects of gamma-rays on acetamide have been of particular interest to the chemists. The modes of radiolysis in both unsaturated and saturated hydrocarbons have also been under investigation.

in life expectancy of animals caused by radiation has been called radiation-induced aging, because the process resembles natural aging. Both kinds of aging are being studied, in order to establish the mechanisms involved and to determine the relationship between the two processes. To test the possibility that aging results from the accumulative effects of nonspecific stresses, noxious agents

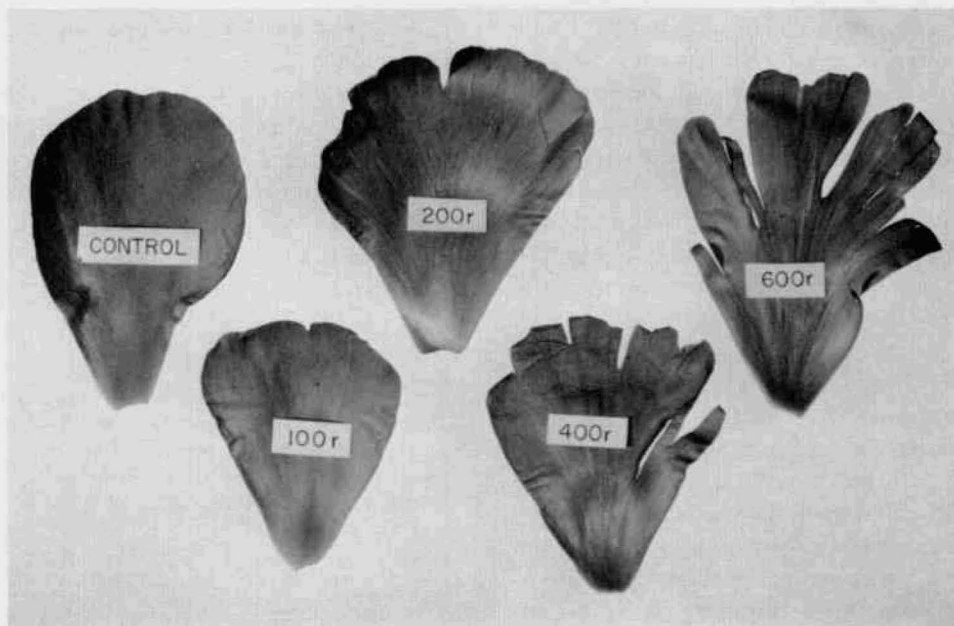


Figure 11. Petals of peony (*Paeonia* var. *Felix Crousse*) showing radiation effects. During dormancy, buds in the early stages of development were exposed to varying x-ray dosages. Eight weeks later the plants were forced to flower. The deep notches probably indicate cell deaths early in petal development, and the light streaks are somatic mutations from red to white color.

Nuclear transformations produce chemical effects whereby atoms recoil from neutron capture. The chemistry of recoiling  $C^{14}$  has been under investigation for several years, and this research is continuing;  $P^{32}$  recoils from thermal neutron capture in solid and liquid phosphorus compounds are also being studied; and the technique of paper electrophoresis is being employed in studies of hot-atom reactions in a series of cobalt complexes.

In investigations on the effects of radiation on living organisms, the methods of molecular biology are receiving increasing attention. Concepts and techniques in this area have found immediate application in such pressing problems as the nature of radiation-induced mutations and of the mechanism of radiation damage in mice. The decrease

such as tetanus toxins were administered to mice in varying doses. These experiments indicate that stress *per se* does not produce a decrease in life expectancy, while radiation differs from other stresses in that it does uniquely shorten life. It may be that radiation constitutes a specific stress on some organ as yet unsuspected. This concept is currently under investigation.

The integrity of certain cell systems, notably the intestinal epithelium, bone marrow, and skin, depends upon continuing proliferation. The radiation syndrome in such systems involves three major questions: (1) how does radiation interfere with cell production; (2) how does the cell population respond to interference with cell production; and (3) what are the consequences to the whole organ-

ism of damage to cell systems? In intensive studies of the effects of radiation on intestinal epithelium, the phenomenon of adaptation has been found to occur under repeated or continuous irradiation.

An especially interesting finding was reported this year by one of the cooperators in the Cooperative Radiation Mutation Program. Researchers at the New York State Agricultural Experiment Station in Geneva have discovered that, as a result of x-irradiation of apple scions and grafting onto unirradiated stocks, the usual chromosome numbers in the stem apex were altered. A variety of new chimeral types have been produced, among them some that may yield the diploid gametes required by the breeding program. Other types indicate that a differential radiation sensitivity, based upon both the chromosome number and the location of the different ploidy levels, has resulted in selective cell destruction followed by regeneration of a new stem apex from the surviving cells.

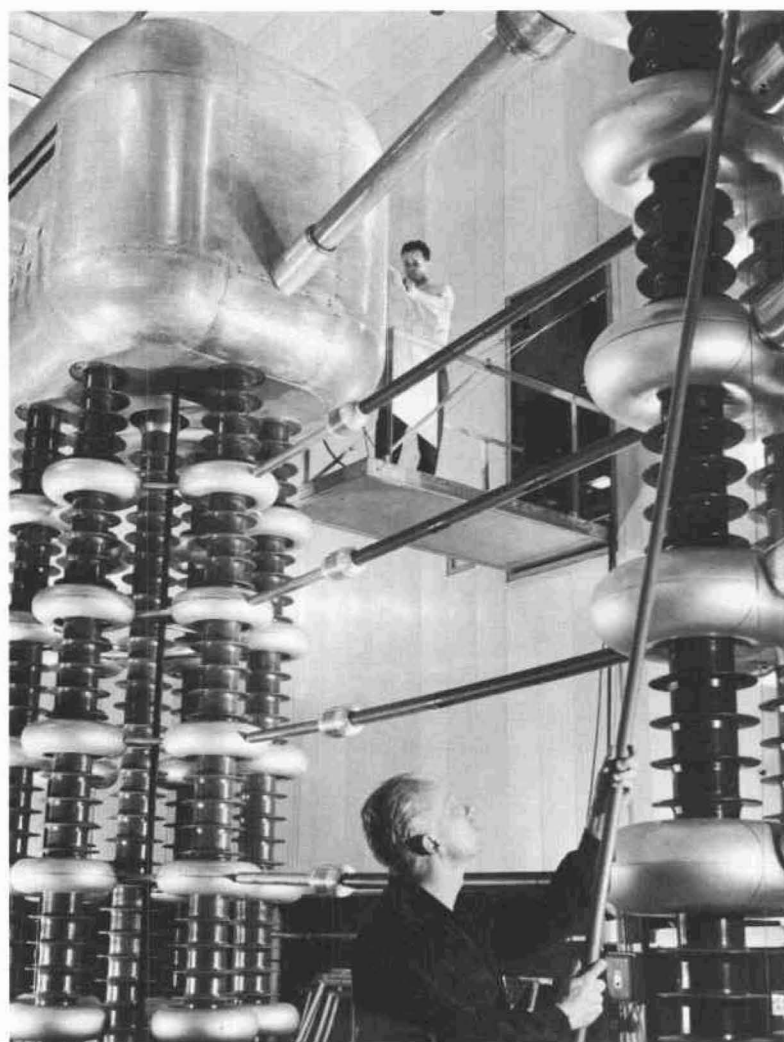
The causes of the great variation in the radiation sensitivity of different plant species has long been a central problem in radiobiology. The results of a ten-year study of several species of higher plants grown under chronic irradiation in the Brookhaven gamma field have now provided information that appears to permit prediction of the tolerance of a given plant to a given exposure of ionizing radiation. More than 200 species representing some 50 plant families have been studied, and analysis of the relationship between sensitivity and nuclear volume indicates that an almost constant number of ionizations per nucleus is required to produce growth inhibition or death. Extrapolation of certain kinds of radiobiological data from one species to another can now be made on a reasonably sound basis; it is suggested that these data might be effectively extended from plants to animal species.

In the Medical Department, the largest single effort has been in the development of neutron capture therapy techniques to control malignant intracranial neoplasms and to study the effects of neutron infiltration upon cellular structures. At the Medical Research Reactor, experiments with rabbits and mice have been conducted to study the effects of intense thermal neutron doses on the head and eye and their use in the control of transplantable tumors. In the treatment of far advanced stages of glioblastoma multiforme, four patients were treated with  $B^{10}$  administration and thermal neutron exposures of from 100 to 200 sec.

Studies of the pharmacology of boron compounds are continuing, but lithium compounds and salts are also being investigated as possible media for neutron capture research.

In addition to the use of reactor neutrons for radiological experimentation, the Medical Department is studying the use of a variety of particles produced in accelerators. With these machines it is possible to produce highly monoenergetic beams and thus obtain uniform dosages. In one experiment 20-Mev deuteron beams from the 60-in. cyclotron are being used to produce lesions in the cerebral cortex of animals. It has been possible by this technique to produce a lesion at a depth not exceeding 2 mm. Damage was found in an isolated strip of cortical tissue of the thickness of only a very few neurons. Another experiment in this field has been conducted by the Biology Department.

Figure 12. The Cockcroft-Walton generator in the Linac Building at the Alternating Gradient Synchrotron. In this machine protons are accelerated to 750 kev; they are then injected into the linac for acceleration to 50 Mev before entering the main AGS magnet ring.



A detailed estimate of the human lethal dose of radiation, including both military and civilian aspects, has been undertaken. This study indicates that the LD<sub>50</sub> for man probably approximates a 350-r tissue dose, under conditions of fairly uniform dose distribution throughout the body and at a fairly rapid dose rate. In another area of research in environmental medicine, possible retardation of growth and development has been reported in children exposed to atomic bomb and fallout radiation; this finding has led to a study of tibial growth in rats subjected to varying doses of radiation.

#### Use of Nuclear Tools

This highly diversified field of research overlaps to some extent the one just described. Only a few of the numerous studies can be mentioned here to indicate the wide range of scientific interests served by nuclear tools of all kinds. Both physicists and chemists have been investigating the arrangements of atoms, ions, and magnetic spin systems as revealed by low energy neutrons from the Graphite Research Reactor, while studies of chemical reactions and of biological processes in plants, animals, and man have been pursued through the use of radioisotopic tracers.

In cooperation with the Chemistry Department of Columbia University, a study is being made at the reactor of slow neutron scattering by hydrogenous solids, liquids, and gases. The total scattering cross sections per proton have been obtained for a number of ammonium halide salts, liquid methylbenzene compounds, and other chemical compounds. By means of recently developed techniques using polarized neutron beams, physicists are attacking several important problems, such as the determination of the *g*-value for various slow neutron resonances, and the collection of data on scattering effects at a wide range of energy levels. With an atomic beam resonance apparatus, nuclear spins and related properties of radioactive nuclei are being measured; another experiment, with optical interference spectroscopy, is directed toward determination of the nuclear spin of Ar<sup>39</sup>.

An organic reaction of interest to chemists for some years is the biosynthesis of nicotine in the tobacco plant. A number of small molecules, labeled with C<sup>14</sup> in various positions, are supplied to root cultures, and the pathways utilized by the root for the synthesis of nicotinic acid are thus determined. The distribution of activity between the

two rings of the nicotine molecule has been traced in the cases of labeled fumaric, aspartic, malonic, and succinic acids and a number of other organic compounds; in addition, there are indications of the ways in which these compounds may be utilized for the synthesis of the pyridine ring.

For some time it has been suspected that the sun is not the major source of cosmic radiation. Evidence recently obtained from studies of cosmic-ray spallation products in meteorites indicates that the radiation flux is fairly constant throughout the solar system. The technique used in such studies is the measurement of the ratio of a short-lived and a long-lived radioactive spallation product in a recently fallen meteorite. The amount of the isotope with the short half-life reflects the cosmic-ray intensity over the last portion of the meteorite's orbit before it intersected the earth's orbit, while the amount of the long-lived isotope indicates the cosmic-ray intensity averaged over the meteorite's entire eccentric orbit. This ratio is then compared to the production rates of these two isotopes, as obtained by bombardment of a sample of the meteorite with 3-Bev protons from the Cosmotron.

A program of study of the composition of  $\approx 200$  specimens of ancient glass is being conducted by means of emission spectroscopy, flame photometry, x-ray fluorescence, and neutron activation analysis. The concentrations of 26 elements in the specimens under study have been determined. The aims of this research are to establish correlations between composition and provenance, date of manufacture, methods of ancient technology, and related questions.

In cell physiology studies, the metabolism of ribonucleic acid (RNA) in dividing cells has been traced by the presence of tritium-labeled cytidine, a precursor of both RNA and deoxyribonucleic acid (DNA). The localization of the precursor within the cell is then determined by autoradiography. It is deduced from this research that, while DNA appears to serve passively as a master template for storing genetic information, RNA serves as the intermediate which transfers the information to sites of protein synthesis. Medical scientists are using a related tracer technique, with tritiated thymidine, cytidine, and uridine as the precursors to the synthesis of DNA, to delineate new hypotheses concerning malignant cell proliferation. The results suggest the existence of a DNA precursor more intricate than a mononucleotide, in a closed system of precursors participating in DNA syn-



thesis. These experiments were conducted on HeLa cells, hemopoietic tissue, and human lymphosarcoma. This research has also been applied to the problems involving antibody synthesis as well as DNA synthesis in embryo and adult mice, in osteoclasts of bone, in fracture repair, and in peripheral blood cells.

In studies on the mechanism of muscular contraction,  $O^{18}$ -labeled water is being used to trace the manner in which the energy source adenosine triphosphate transfers its energy to the contractile protein actomyosin. Other studies in biochemistry concern the molecular structure of the enzyme ribonuclease; the involvement of the many enzymes of green leaves in photosynthesis, in respiration, or in both; and the effect of radiation in inducing chemical modification of the side chains of enzymes and other proteins.

The basic biological problem of membrane permeability is being studied with isotopic tracers, and the testing of certain older theories by this method has raised many new questions. Criteria for the evaluation of data from isotope experiments have been developed, and it has been found by theoretical investigation that the description of the transfer process through membranes derived by concepts of statistical mechanics can be more easily attained by the application of Newtonian mechanics.

In conducting analyses for trace metals occurring in very small concentrations in serum, plasma, and blood, medical scientists have selected neutron activation analysis as the most sensitive method. This technique consists of exposing the sample to neutron irradiation in the reactor, which induces radioactivity in the various elements composing the sample. Then, to measure any of the many elements, it is necessary either to sort out the radiations characteristic of each, or to separate by chemical methods the elements of interest. Both approaches are now in use in the Medical Department. The first requires suitable electronic circuitry to favor the "interesting" radiations, such as that from manganese, which is an ultramicro-constituent of tissues. The second approach has also been developed into a clinically useful micro-procedure, so that the plasma or serum previously required in large samples can now be analyzed in fractions of a milliliter. The technique of neutron activation has been applied to the precise location in the brain of the satiety center. In this experiment animals were injected with gold thioglucose,

which induces a tendency to overeat. Neutron activation of gold and sulfur in brain slices from such animals revealed the precise location and other structural features of the satiety center.

Vasopressin has long been implicated in hypertensive phenomena in animals and man. It has now been possible to synthesize a highly radioactive form of this hormone and thus to elucidate important facts concerning its functional attachment to its receptors. The evidence pertaining to vasopressin has suggested approaches to the study of insulin and other hormones that may be useful in the interpretation of their physiological action.

In studies of ketogenesis in man, the use of  $C^{14}$ -labeled compounds has provided considerable information regarding the biochemistry and pharmacology of diabetes. These studies have extended the knowledge of glucose formation in man in terms of its metabolic pathways, its precursors, and its breakdown products. The regulation of lipid and cholesterol metabolism by hormones was also studied with similar techniques.

Studies of patients with multiple myeloma have revealed significant metabolic differences between various types of the disease with respect to the metabolism of normal and aberrant proteins. This research has been greatly assisted by the use of total-body counting techniques, as have studies of  $Co^{60}$ -labeled vitamin  $B_{12}$ . In other studies of multiple myeloma, tritiated thymidine has been used to determine the proportion of myeloma cells retaining their capacity to propagate the tumor; from this it appears that only a small number of the cells are involved, while the balance reach a mature state and remain as parasites to the host organism. These observations might also be applied to the leukemias and to solid tumors currently under investigation.

#### **Research Related to Nuclear Energy Development**

In this category of scientific activity at Brookhaven are theoretical and experimental investigations of the neutron scattering and absorbing properties of substances used in nuclear reactors and atomic weapons, general studies of breeding reactors, attempts to resolve uncertainties in reactor theory, and evaluation of new reactor concepts in terms of nuclear physics. Research of a more specific nature is concerned with economic evaluation of reactor concepts, studies of the oxidation and corrosion of reactor components, development of radiation applications, investigation of better

methods for reprocessing nuclear fuel and disposing of nuclear waste, and development of reactor components.

Studies at Brookhaven are contributing to the refinement of reactor physics theory and eventual progress in reactor design technology. New light is being cast on neutron thermalization, resonance absorption of neutrons, fast fission, nonlinear reactor kinetics, and chemical binding effects. Under study is reactor stability against xenon poisoning, of concern in reactors operating at high power and high flux levels. In a study of thermal breeding it was shown that the Th- $U^{233}$  cycle is feasible with solid as well as fluid fuels, provided that a weakly absorbing moderator is used, and that fast fission is important in improving the breeding ratio in such systems.

At the request of the AEC, Brookhaven is implementing a program for continuous review and maintenance of cross section data. Files of data and cross section codes have been assembled, and measurements have been made to provide cross section values representing a best available adjustment between theory and measurement.

To obtain a better knowledge of the properties of the  $U^{238}$  nuclide, a quantitative evaluation of the resonance absorption of neutrons in  $U^{238}$  has been achieved by using improved experimental measurements of resonance parameters and powerful analytical techniques such as Monte Carlo calculations. A new and better technique has been developed for measuring  $\rho_{28}$ , the ratio of  $U^{238}$  captures above and below the cadmium cutoff. Values of  $\delta_{28}$  and  $\epsilon$  have also been measured by an improved technique using water lattices with uranium metal rods. Miniature rod lattices have been used to confirm and improve earlier determinations of the fast fission factor and resonance escape probability.

In addition, exponential experiments in water with plate and rod lattices were performed to investigate the effect of the geometry of loading on the buckling deduced from flux traverses. In the past, theoretical analysis of lattice experiments in water has been difficult because of uncertainties in chemical binding effects when the free gas model is used. These uncertainties have been eliminated through use of improved models for neutron thermalization, and the discrepancy between theory and experiment has been reduced.

Better knowledge is needed of the properties of graphite relevant to nuclear physics. By using the

results of separate experiments with pulsed neutrons and static diffusion, neutron diffusion parameters were determined. In addition, an anisotropy in neutron diffusivity was revealed, which had its maximum value in the direction of the axis of extrusion of the graphite. An experimental value of the neutron slowing-down time in graphite was determined and compared with a calculated value obtained from a rapidly converging series solution of the time-dependent Boltzmann equation with constant scattering cross section.

To test the validity of the assumption, recently questioned, that asymptotic reactor theory is valid in bare homogeneous assemblies, an experimental facility has been constructed in which perturbation from reflected neutrons is at a minimum; hence it should be possible to make independent determinations of both critical mass and buckling.

In connection with the design of the High Flux Beam Research Reactor,  $\approx 700$  critical experiments were performed to determine the effect of various geometrical configurations upon reactivity and to assist in arriving at an optimum design. Completion of so many critical experiments in a short time was possible only through use of several new tools and techniques, including a poisoning method for measuring excess reactivity of highly supercritical assemblies and an analogue computer programmed to accept a flux signal from the critical assembly and instantaneously solve the reactor kinetic equations to obtain reactivity.

Assistance given to the AEC in the evaluation of specific reactor proposals, as part of a program initiated in December 1957, included consultation on the safety aspects of the Consolidated Edison Thorium Reactor and evaluations of the NS Savannah project, the proposals for Euratom, and the spectral shift reactor. Other projects included the economic appraisal of various chemonuclear systems employing fission fragment energy for the fixation of nitrogen; a survey of the status and potential of direct conversion of fission energy to electrical energy without conventional thermal means; and investigations of reactors employing nonrigid fuels such as fluidized solids, intermetallic dispersions in fixed fuel elements, and granular composites of moderator and fuel. A detailed transfer function analysis of the Liquid Metal Fuel Reactor Experiment demonstrated the inherent stability of this reactor concept. The effect of different concrete shielding materials on the attenuation of delayed neutrons from fissions was determined.



The behavior of construction materials in different reactor system environments comprised a large segment of metallurgical research. Environments studied included liquid metals, slurries, fused salts, radiation fields, temperatures  $>2000^{\circ}\text{C}$ , and oxidizing and reducing atmospheres. Specific research programs were concerned with both heat transfer and electrochemical types of corrosion, with corrosion inhibition, erosion, oxidation, irradiation damage and annealing, and temperature-induced changes in physical properties. These studies required static experiments, dynamic experiments with circulating metal and circulating and agitated slurries, thermodynamic measurement with galvanic cells, spectroscopy of fused

salts, high temperature measurement, and physical and mechanical measurement. The many unusual problems encountered necessitated the development of new methods for measurement and observation to supplement the more conventional metallurgical techniques.

Radiation chemistry and radiation engineering are two important areas of applied research in nuclear energy. At Brookhaven research in applied radiation chemistry has been concerned largely with radiation-induced polymerizations, including solid state, graft, and ionic polymerization. Electron spin resonance has been used to study the nature and lifetime of free radicals in nylon. Research has been started to serve as a basis for a



Figure 13. Bubble chamber film is given careful inspection for all nuclear interactions of interest to physicists. These machines, known as scanning tables, project an image of the bubble chamber film enlarged to actual size, so that each frame may be inspected easily. Scanning is the first in a series of operations designed to extract information from the film.

theory of radiation effects in substituted aromatic compounds. The radiation-induced chemical synthesis of economically important nitrogen compounds was studied, and preliminary experimental data were obtained.

In the field of applied radiation engineering, experimental and theoretical studies have been made of large-scale, low level irradiators and various source configurations in high level radiation facilities. The AEC exhibits in New Delhi, India, and Cairo, Egypt, included radiation facilities provided by BNL and operated by BNL personnel. A food irradiation research facility is being built to serve as a prototype of facilities to be installed at several research centers throughout the country. A high level source development laboratory has been designed and its construction in 1961 has been approved.

The many types of nuclear reactor fuel elements now used or proposed for use in military and civilian power reactors have complicated the problem of reprocessing used fuel. At Brookhaven problems in existing fuel reprocessing operations are being studied, and new fuel reprocessing systems employing nonaqueous methods for the dissolution, volatilization, and separation of fuel element components are being developed. In one process gaseous reagents are used in a fluidized bed of inert solids. In another, the dissolution of spent fuel elements in a new, anhydrous, inorganic solvent medium is followed by volatilization, separation, and recovery of the fissile components with known methods. In this process interhalogens can be safely employed in the volatilization, separation, and recovery steps, although their use in the dissolution of uranium metal has been shown to be potentially hazardous.

Work has also been initiated on aqueous fuel reprocessing methods for the recovery of uranium from the types of fuel used in the more advanced designs of naval reactors; several methods were investigated for the preparation of tetravalent uranium solutions, for reduction of plutonium in the Purex process partition cycle; and experimental work was begun on amalgam reduction of  $\text{UO}_2$ .

The processing and disposal of waste is an important phase of fuel cycle development. At Brookhaven, research in this field has been centered mainly on isolation and concentration of the long-lived fission products either by solvent extraction techniques or by absorption and fixation on mineral exchange material. Other research is con-

cerned with the calcination of certain liquid waste to form a stable solid.

Research on reactor components included a search for liquid and gas coolants with good resistance to radiation; investigation of bearing materials for liquid metal service; testing of a large liquid bismuth flow system incorporating commercial scale valves, piping, heat exchangers, and pumps in a radiation-free environment; and establishment of a radiation loop to ascertain the effect of reactor neutron irradiation on a piping system in which corrosion-inhibited uranium-bismuth fluid is circulated. A control loop, the radiation-free counterpart of the radiation loop, was operated for 3000 hr to provide a standard against which to measure the effects of reactor irradiation. Because of the curtailment of the Liquid Metal Fuel Reactor project in February 1959, research in progress was terminated, except for experiments of long duration. The last of these is expected to be completed in 1961, when the radiation loop will be withdrawn from the reactor and subjected to metallurgical examination in the High Level Metallurgical Hot Cell built for this purpose.

In power reactors, as in conventional power generators, the conversion of heat to useful power can be made more efficient if the heat source is operated at a higher temperature. In connection with gas-cooled high temperature power reactors employing graphite-encased or graphite-uranium composite fuel elements, the diffusion of xenon in high density graphite is being studied. The continuous absorption of xenon from a helium gas stream is also being studied to develop information for the design of purification systems. Investigation has been started of graphite-encased and graphite-uranium composite fuel elements. This program necessitated the development of a new thermocouple element for use at temperatures above  $2000^\circ\text{C}$  in a reducing atmosphere.

Because of increasing interest in the use of molten metal heat transfer media in proposed designs of high temperature power reactors, additional information is needed on the effect of metal flow conditions upon the heat transfer coefficient. Predictive equations were developed analytically for conditions of turbulent parallel flow through unbaffled tube banks, and the results were compared with data from experiments designed to test the validity of the equations. Additional experimental work is planned to resolve discrepancies in the results.

## **Development and Practical Application of Research Tools**

This, the last of the five categories of scientific activity at Brookhaven, includes the design and development of special research tools such as accelerators and reactors and their ancillary technical equipment. With the exception of the 60-in. cyclotron and the electrostatic generator, the Laboratory is responsible for the development, design, and construction of all the facilities described under the headings "Present Research Facilities" and "Additional Research Facilities." The preparation of special isotopes and the development of radiation sources also come under this heading.

Brookhaven scientists, almost from the inception of the Laboratory, have been among the leaders in the development of new particle accelerators and of the external beams, detectors, bubble chambers, and electronic instruments that have so greatly enhanced the value of these tools for fundamental research in physics.

After several years of concentrated effort on the design and construction of the 30-Bev AGS, the construction phase was substantially completed during fiscal 1960, and the design and procurement of components for experimental beams at the AGS was begun. The design of circular and rectangular quadrupole magnets, of bending magnets, targeting equipment, and special straight sections has been completed. Most of the 43 magnets required for the AGS have been ordered, and some have been built. A beam separator housed in a vacuum tank is being designed as a module that can be coupled with similar modules to form separators of different lengths. Specifications and drawings have been completed for modification of the AGS Target Building to accommodate bubble chambers using explosive gases.

Despite the concentration of effort at the AGS, developments leading to improved operability and utilization of the 3-Bev Cosmotron have not been neglected. With the addition of a second Piccioni ejection magnet system at the Cosmotron this year, three external beam paths of high energy protons can now be used in the new experimental area. These external beams, extracted with an efficiency of  $\approx 25\%$ , can be focused and analyzed to provide separate proton or pion beams of various energy levels up to  $\approx 3$  Bev. A major advantage of the use of external beams, as contrasted with the use of the internal circulating beam, is that an experiment can be prepared for one beam while an-

other is in progress at a second beam. Thus, full utilization of the Cosmotron's running time is now possible.

The external beams are extremely effective when used in conjunction with bubble chambers to probe particle interactions and strange particle production. Development has been rapid since the bubble chamber was invented in 1952. By 1955, BNL had developed a 2 $\times$ 3-in. bubble chamber, which was considered large. Today, 20-in. bubble chambers are commonplace. As particle beams of increasing energy become available, even larger bubble chambers will be needed. For this reason an 80-in. hydrogen bubble chamber is being designed for use with the AGS.

In operating a bubble chamber, three or four photographs are taken simultaneously at a predetermined instant following the release of pressure on the superheated liquid. These stereophotographs record the position of bubbles appearing on, and thus defining, the ionization tracks associated with the passage of charged particles through the liquid. An analysis of each set of stereophotographs yields three-dimensional space coordinates for the trajectories of incident charged particles and of charged particles emitted from interactions between incident particles and nuclei in the liquid. Data from many such analyses can be used to determine, for specific particles, properties such as mass and charge, cross sections of interaction, average lifetimes, and modes of decay.

Because of the infrequent occurrence of certain events of interest to the scientist,  $\approx 100,000$  stereophotographic sets may be required for statistical treatment of a single bubble chamber experiment. In exploratory experiments at the Cosmotron during the past year, 30,000 exposures were made with the Cambridge 20-in. propane-methyl iodide bubble chamber; 70,000 exposures of negative pion beams at 1 Bev were made with the Columbia 12-in. hydrogen bubble chamber; and the BNL 20-in. hydrogen bubble chamber was used to study proton-proton interactions at 2 Bev in an experiment that produced 50,000 exposures. The magnitude of the problem of reducing such photographic records to useful data is apparent when it is understood that thousands of photographs must be scanned to select events of interest, and that these events must then be examined microscopically to obtain coordinate data from which to compute trajectories.

This description of the analysis of bubble chamber events illustrates the growing problem of data

handling. The approach to solution of the problem at Brookhaven is to develop automatic systems for data taking, time and amplitude sorting, automatic plotting, and similar activities, as well as machine methods for data reduction and computation. Some experiments are so complicated that they can be carried out only with the use of highly automatic data handling circuits. In other cases the instrumentation provides more reliable results or makes possible a saving in manpower.

Two relatively elementary devices have been developed as an initial step in automation of the tedious process of analyzing bubble chamber photographs. The special microscopes used for examination of the bubble chamber photographs have been equipped with systems for punching the coordinate data onto IBM cards in digital form. An electrochemical device now being used with these microscopes provides automatic determination of gap-length distributions of bubbles along particle tracks. The distribution of bubbles is a function of the specific ionization of the charged particle; hence the specific ionization can be computed from such data. It is probable that instrumentation can be developed to scan photographs automatically, select events of interest, and record the particle track coordinate data without human intervention.

Experiments in high energy physics also involve complex electronic devices such as detectors, discriminators, pulse amplifiers, counters, and circuits for time measurement at very high pulse rates. Such devices must be capable of operation with pulses of a few nanoseconds' duration ( $10^{-9}$  sec), approximately the time required for light to travel one foot. Transistor circuits for such application are now being designed to serve as basic "building blocks" of compatible components that can be interconnected as required for use in a specific experiment. Available building blocks now include a multicoincidence circuit, a pulse amplifier, a fast scaling circuit, and a "fan-out." Each building block is in a small chassis, with the necessary connectors and controls, so that a substantial number of them may be mounted on a supporting structure and connected with ease. Work is continuing on the design of new building blocks and the improvement of those now in use.

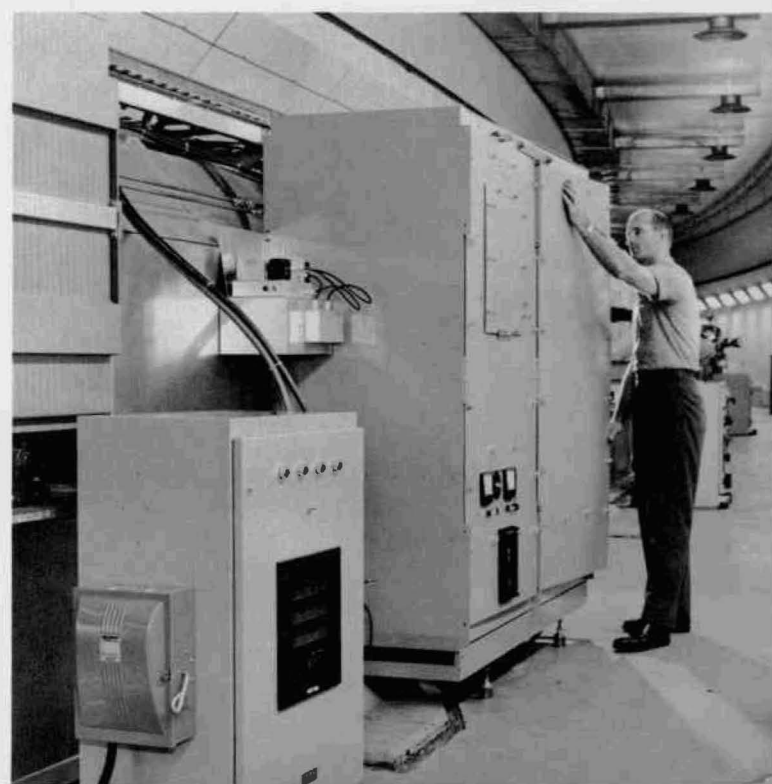
The solid state detector, similar to a junction transistor, is an important addition to the family of radiation detectors because of its very high energy resolution with heavily ionizing particles, its

linear energy relationship, and its fast speed of response. Bell Telephone Laboratories, the inventors of the junction transistor, invited Brookhaven National Laboratory to cooperate in a study of solid state detector devices; arrangements were made for BNL to supply electronic equipment and measure the performance of the detectors. To date some excellent experimental detectors and several other interesting solid state devices have been produced in this joint venture. A preamplifier having low noise and high gain was designed for this specific application.

Other electronic devices built for use as research tools include an apparatus for chemical analysis by automatic derivative polarography, an analogue computer for rapid analysis of data on electrolyte exchanges in human beings, a reactor modulator and analogue tape system for reactor physics experiments, a precision regulator circuit for Cosmotron magnet power supplies, precision regulated power supplies for a beta-ray spectrometer, a generating voltmeter readable to a fraction of a kilovolt for the 4-Mw electrostatic generator, and a modulator for a smaller electrostatic generator in the Chemistry Department.

Crystal spectrometers continue to be used at the Brookhaven Graphite Research Reactor to pro-

Figure 14. One of the 12 radio-frequency accelerating stations for the Alternating Gradient Synchrotron. The smaller cabinet contains the control and auxiliary circuits for the rf station. The larger cabinet contains in its left portion the rf power amplifier, which provides the drive for the ferrite accelerating cavity just visible behind it; and in its right portion, the saturating supply, which tunes the ferrite cavity as the frequency of the accelerating voltage is raised.



duce highly monoenergetic beams of slow neutrons and to analyze in fine detail the energy distributions of slow neutrons in cross section measurements. Techniques developed under a continuing program of spectrometer improvement have made available monoenergetic neutron beams of any desired energy between 0.0006 and 10 ev.

In a research program to determine the  $g$ -value for various slow neutron resonances, the use of polarized nuclei as targets for polarized monoenergetic neutrons has been proposed. To obtain the low temperature required for production of polarized nuclei it was necessary to develop cryogenic techniques capable of maintaining temperatures close to absolute zero for time periods of the order of hours. A cryostat using two stages of adiabatic demagnetization was developed in which heat leaks as low as 25 ergs/min have been observed at  $0.01^\circ\text{K}$ ; the temperature remained below  $0.02^\circ\text{K}$  for more than an hour. Even better performance is anticipated from the use of special techniques for preparing paramagnetic salts and the adoption of innovations in the salt supporting structure and in the design of the thermal valve heat switch. Another phase of the program resulted in the development of a polarization spectrometer that provides a neutron beam with polarizations of  $>98\%$  and has a magnetic guide field arrangement for selectively rotating the direction of polarization without appreciable depolarization.

Since 1950 the Laboratory has been greatly interested in the utilization of intense radiation sources, the development of such sources, and the utility and availability of radioisotopes in general. Before widespread practical application of radiation sources in industry becomes possible, engineering information on source technology must be developed and made available. The design of a radiation development laboratory is almost completed, and construction should begin in the coming year. In this laboratory it will be possible to investigate the hazards inherent in megacurie sources, to study problems of heat generation and dissipation in high level sources, to conduct radiation damage studies of source materials, to continue the development of high level dosimetry systems, and to develop methods of handling, encapsulating, and testing high level sources. An irradiator for use in food irradiation studies at several research centers has been designed, and a prototype employing  $\approx 25,000$  curies of  $\text{Co}^{60}$  is being built.

The general purpose of isotope development is to increase the utility and availability of radioisotopes. The direct objective is to develop methods of producing isotopes that would otherwise be unavailable, or to put available isotopes into more usable forms, for example, by increasing specific activity or isotopic purity. The Laboratory has made available carrier-free isotopes of short half-life through a technique based on isolation and shipment of the parent fission product. The short-half-life daughter isotopes  $\text{I}^{132}$ ,  $\text{Tc}^{99m}$ , and  $\text{Y}^{90}$  may be separated by simple "milking" systems from the parent material. Other isotope separation processes being developed include methods for producing  $\text{Ca}^{47}$ ,  $\text{Cu}^{67}$ ,  $\text{I}^{124}$ ,  $\text{Ga}^{68}$ ,  $\text{Sc}^{47}$ , and  $\text{Kr}^{83}$ . Production of  $\text{Ar}^{38}$ , of interest as a calibration standard for mass spectrographs, has begun. Modifications have been made in existing processes to reduce  $\text{H}^3$  contamination in  $\text{Mg}^{28}$  and to improve the yield of  $\text{Te}^{132}$ . Separation of  $\text{Sm}^{151}$  from aged fission product solution is now being done on a larger scale.

#### ADMINISTRATION

The organization of the Laboratory as of June 30, 1960, is given in Figure 1. Responsibility in the Director's Office for the Laboratory's service and administrative divisions was transferred to Mr. G.F. Tape and Mr. S.M. Tucker following the departure of Mr. W.H. Fields, Jr., in February 1960. The Personnel Division reports directly to Mr. Tape, the Deputy Director. The Supply and Materiel and Purchasing Divisions also report to Mr. Tape; however, responsibility for coordination and supervision of their work has been assigned to Mr. E.C. Smith. The Architectural Planning, Plant Maintenance, Central Shops, Security and Plant Protection, Information, and Photography and Graphic Arts Divisions report to Mr. S.M. Tucker.

In May 1960 the Physics Department was expanded to include members of the Cosmotron Department who are engaged predominantly in research proper. Dr. M. Goldhaber was appointed Chairman of the enlarged department, with Dr. S.A. Goudsmit and Dr. R.L. Cool as Deputy Chairmen, the latter having specific responsibility for coordinating the high energy experimental research program. Plans were also announced for combining the Accelerator Development Department and the Cosmotron Department into a new Accelerator Department under the chairmanship of Dr. G.K. Green in the fall of 1960.



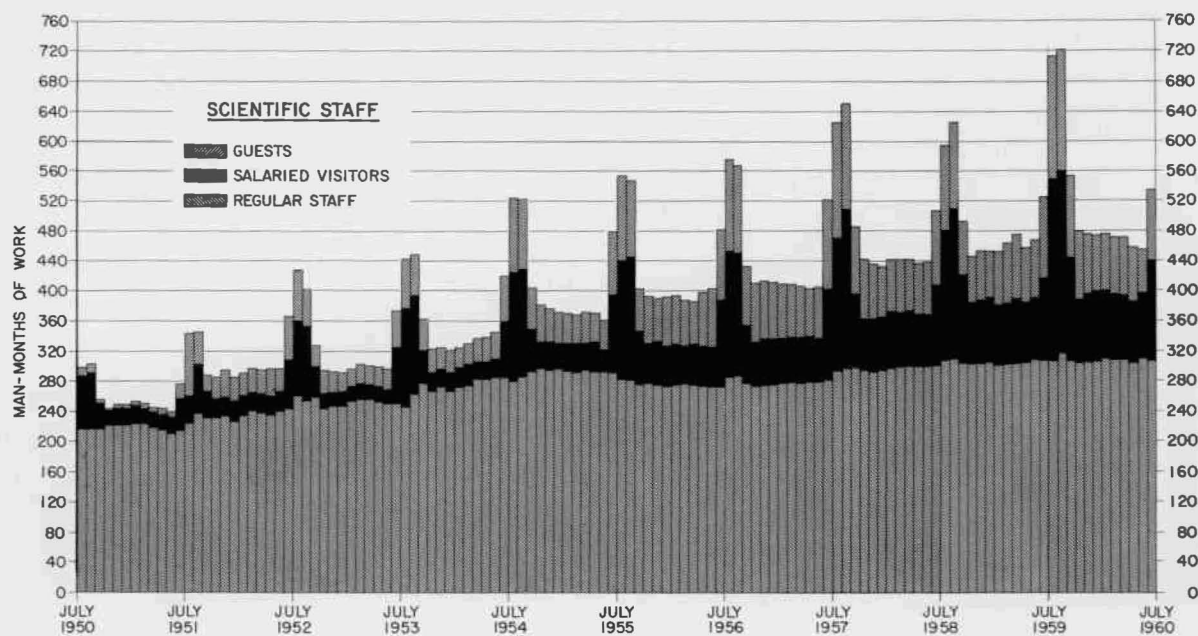
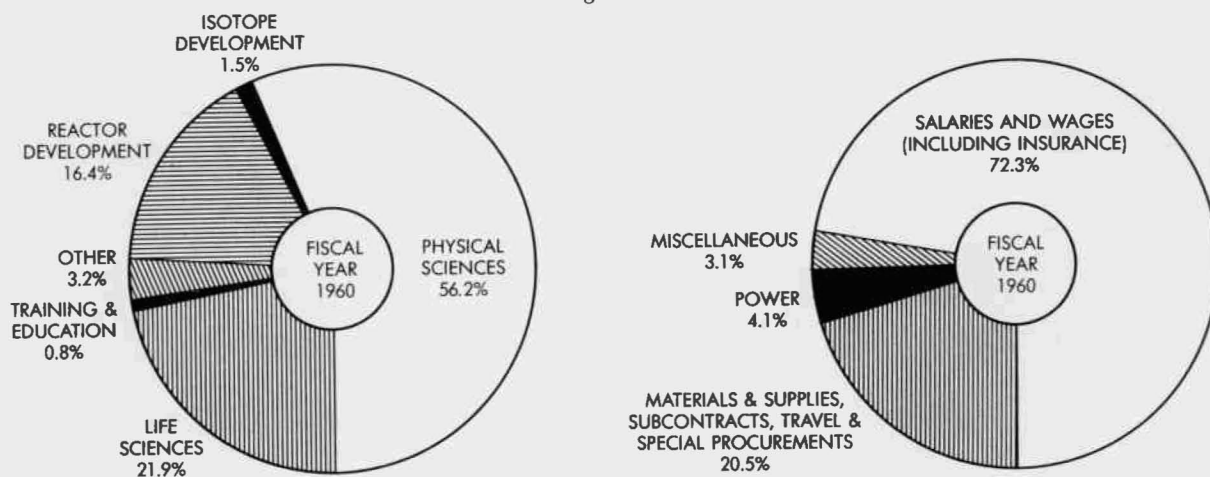


Figure 15.



OPERATING COST DISTRIBUTION BY PROGRAMS

FISCAL YEAR	PHYSICAL SCIENCES	REACTOR DEVELOPMENT	LIFE SCIENCES	ISOTOPE DEVELOPMENT	TRAINING & EDUCATION	OTHER (NET)	TOTAL COST
1960	11,775,522	3,421,837	4,584,337	315,310	166,300	671,205	20,934,511
1959	9,395,450	4,410,595	4,172,833	104,486	39,442	538,498	18,661,304
1958	8,281,717	5,114,706	3,609,035	26,086	98,030	577,726	17,707,300

MAJOR CATEGORIES OF OPERATING EXPENDITURES

FISCAL YEAR	SALARIES AND WAGES	MATERIALS & SUPPLIES SUBCONTRACTS, TRAVEL & SPECIAL PROCUREMENTS	POWER	MISCELLANEOUS (NET)	OPERATING TOTAL
1960	15,125,492	4,295,397	868,432	645,190	20,934,511
1959	13,248,663	4,220,091	728,766	463,784	18,661,304
1958	11,381,555	4,841,907	742,006	741,832	17,707,300

Figure 16.

## FINANCE

The Laboratory's AEC-supported research was financed by five Divisions of the AEC, namely, Reactor Development, Research (Physical Sciences), Biology and Medicine (Life Sciences), Isotope Development, and Training and Education. The operating costs as distributed to these Divisions are shown in Figure 16; the major cat-

egories of operating expenditures are also shown in this figure. Table 1, on the reverse side of the organization chart (Figure 1), shows in detail the operating expenditures on a broad organizational basis.

Table 2 gives the expenditures for capital equipment. A summary of expenditures for fixed assets (plant and equipment) is presented in Table 3. Table 4 sets forth the inventories for which the Laboratory is responsible.

Table 2

### Capital Equipment Expenditures (Including Charges From Organizational Units; See Table 1)

	FY 1960		FY 1959		FY 1958	
	\$	%	\$	%	\$	%
Scientific & hospital	2,760,196	85.2	1,597,742	71.3	955,623	65.0
Automotive & heavy mobile	201,234	6.2	267,452	11.9	186,990	12.7
Office machines & furniture	75,971	2.3	86,840	3.9	85,646	5.8
Shop equipment	142,141	4.4	154,332	6.9	150,668	10.3
Miscellaneous	62,842	1.9	133,195	6.0	91,262	6.2
Expenditures, Total	3,242,384	100.0	2,239,561	100.0	1,470,189	100.0
Proceeds from sales	(39,958)		(23,165)		(27,997)	
Expenditures, Net	3,202,426		2,216,396		1,442,192	

Table 3

### Costs Incurred for Fixed Assets (Including Charges From Organizational Units; See Table 1)

	FY 1960			FY 1959			FY 1958		
	Man-years			Man-years			Man-years		
	Costs, \$	Sci.	Others	Costs, \$	Sci.	Others	Costs, \$	Sci.	Others
<u>Alternating Gradient Synchrotron</u>									
Direct									
Salaries, wages, insurance	1,248,309	28.0	127.0	1,076,228	34.5	112.5	748,473	30.5	77.0
Materials, construction, etc.	2,249,780			6,859,692			4,897,065		
Subtotal direct	3,498,089			7,935,920			5,645,538		
Charges from organizational units	253,025			198,595			162,269		
Total	3,751,114			8,134,515			5,807,807		
<u>Other, Including High Flux Beam Research Reactor and 80-in. Bubble Chamber</u>									
Direct									
Salaries, wages, insurance	277,001	9.5	16.5	147,614	6.5	12.0	98,035	4.0	8.0
Materials, construction, etc.	4,446,120			4,223,724			1,924,177		
Subtotal direct	4,723,121			4,371,338			2,022,212		
Charges from organizational units	160,256			134,974			90,932		
Total	4,883,377			4,506,312			2,113,145		

Table 4  
Inventory at Close of Fiscal Year

Type of inventory	Fiscal year		
	1960	1959	1958
General stores*	\$ 448,487	\$397,842	\$317,906
Precious metals and radium	87,668	73,621	74,128
Stable isotopes	23,413	26,494	29,134
Heavy water	679,535	443,820	—
Total	\$1,239,103	\$941,777	\$421,168

\*The number of months investment was 3.2 in 1960, 3.4 in 1959, and 3.3 in 1958.

BROOKHAVEN NATIONAL LABORATORY  
COMPARATIVE BALANCE SHEET

Assets	June 30, 1960	June 30, 1959
Cash	\$ 681,682	\$ 349,183
Accounts receivable	84,488	151,032
Inventories	1,294,527	953,412
Advances and prepaid items	82,038	379,135
Deposits	57,049	214,458
Fixed assets (less reserves of \$28,824,428 at June 30, 1960, and \$25,366,196 at June 30, 1959)	58,244,540	45,381,296
Construction in progress	21,082,874	17,931,378
Total assets	<u>\$81,527,198</u>	<u>\$65,359,894</u>
Liabilities		
Accounts payable	\$ 2,699,885	\$ 1,609,045
Accrued payroll	104,969	171,795
Atomic Energy Commission	78,722,344	63,579,054
Total liabilities	<u>\$81,527,198</u>	<u>\$65,359,894</u>

NOTE: Although the Laboratory has custody and use of the assets shown on the Balance Sheet, title remains vested in the United States Government.

### CONFERENCES AND PUBLIC INFORMATION

Three formal conferences were conducted at the Laboratory during the year:

1. Symposium on Bioenergetics, jointly sponsored by the Division of Biology and Medicine of the AEC and BNL, October 12-16, attended by 147 scientists representing 75 institutions;

2. Fourth Conclave on Nuclear Energy in Medicine, on the theme, Nuclear Medicine in Surgical Research and Practice, jointly sponsored by the Division of Biology and Medicine of the AEC and the BNL Medical Department, October 26-27, attended by 66 chairmen of departments of

surgery from American and Canadian medical schools; and

3. Thirteenth Annual Brookhaven Biology Symposium, on Protein Structure and Function, June 6-8, attended by 193 scientists representing 70 institutions.

The Seventh Annual Naval Reserve Nuclear Science Seminar, on The Management of Radiation Accidents, was held at the Laboratory September 13-26 and was attended by 33 reserve officers.

A Summer Institute, sponsored by the American Society of Engineering Education, was held August 6-14. The 32 participants, who had al-



ready received a five-week lecture course at Cornell University, received practical indoctrination in various aspects of nuclear engineering, including reactor operations.

In September, the George B. Pegram Lectureship was inaugurated. Named in honor of the late eminent physicist who played a leading role in the founding of the Laboratory, this annual series of public lectures was established to honor an individual who has made outstanding contributions in his field, and to provide a forum for him to discuss the broad implications of science in the area of his experience and interests. The first series, entitled "An Introduction to Space," was presented by Dr. Lee Alvin DuBridge, President of California Institute of Technology. The four lectures, held in the Laboratory's Theater on September 14, 16, 18, and 21, were attended by  $\approx 3000$  persons. Subsequently, the lectures were published in book form by Columbia University Press.

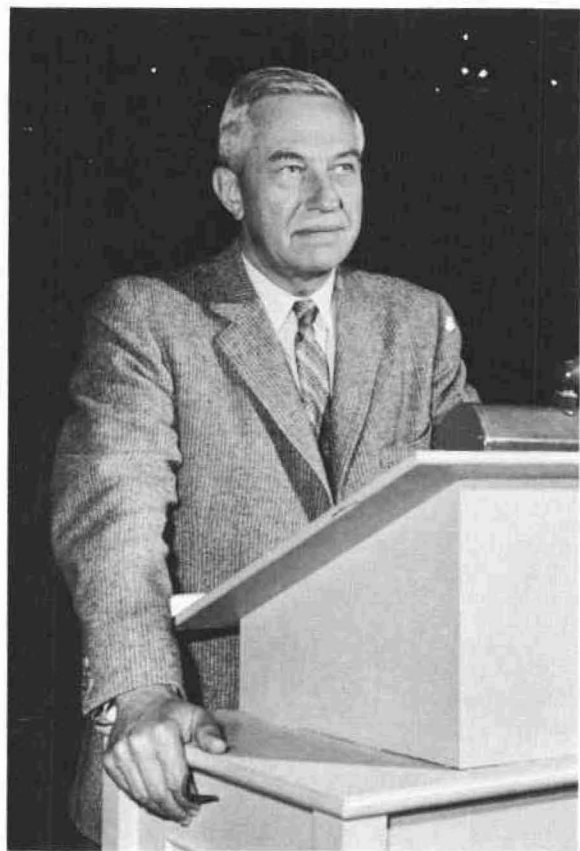


Figure 17. Dr. Lee A. DuBridge, President of California Institute of Technology, speaking during the first Pegram lecture series, entitled "An Introduction to Space."

During the fiscal year, 1193 persons representing 47 professional and technical groups paid one-day visits to the Laboratory to view its facilities and to learn about its research program. Additional visitors included 422 high school and college teachers receiving special training in the nuclear sciences at summer institutes sponsored jointly by the National Science Foundation and the AEC. Eighty-eight graduate students made brief visits to discuss with individual BNL scientists problems in their fields of interest.

Fifty-eight foreign countries sent 904 professional, governmental, and industrial representatives for short visits during the year. In addition, seven foreign research teams came for technical information, and special programs, lectures, and tours were planned for them. The AEC arranged visits for two groups of USSR scientists, under the exchange program sponsored by the National Academy of Sciences of the U.S.

On October 31, 7852 individuals attended the tenth annual Visitors' Day at BNL. On October 23, the second annual College Student Visitors' Day for junior and senior college classes was held; 1034 students attended, representing 65 colleges and universities. On October 24, the sixth annual High School Student Visitors' Day was held; 4143 students attended, representing junior and senior classes from 148 schools.

On February 11, BNL was host to 239 selected high school science students and editors of school papers from 27 Suffolk County schools, as part of the fourth AEC-sponsored Science Youth Day commemorating the birthday of Thomas Alva Edison. This year a new type of program was planned; instead of tours of the Laboratory's major facilities arranged for the entire group, discussion sessions on 20 research projects were set up, and each student attended two sessions of his choice.

Staff members responded to 41 requests to address organizations of laymen and semiprofessional audiences in the vicinity of the Laboratory.

The normal activities of the Public Information Office have continued to increase, mainly because of the heightened interest in the nuclear sciences shown by the public and by students. The growing importance of nuclear energy in the schools' curricula is reflected in requests from students for information and literature. Loans of technical motion pictures from the BNL film library reached an all-time high during the past winter months.

PHYSICAL  
SCIENCES  
AND  
ENGINEERING



# Physics

The research program in physics consists of experimental and theoretical studies concerned with the structure and fundamental properties of matter. Various features of the complex structure and properties of matter can be most effectively studied by observing the interactions of charged particles, neutral particles, and radiation with matter. Some of these studies deal with the interactions involving individual atoms, atomic nuclei, or nucleons, and others with conglomerates of atoms and molecules in bulk matter. The Cosmotron, 60-in. cyclotron, Van de Graaff accelerator, 18-in. cyclotron, and the Graphite Research Reactor at Brookhaven provide a wide range of energies and diversity of particles and radiation with which to carry out experimental investigations. The researches reported here will be described under the categories of particle 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 scientists and provide stimuli for new approaches to the problem.

The work of Brookhaven scientists is enhanced by the presence of a number of visiting and guest scientists on leave from other institutions in this country and abroad. These visitors and guests are attracted to Brookhaven not only by the availability of its facilities but also by the opportunity to collaborate with Brookhaven scientists. The cross-fertilization of ideas and experience resulting from the presence of these visiting scientists is very important to an active research program.

## PARTICLE PHYSICS

### The $\pi$ -Nucleon System

The interactions that take place between nucleons, between nucleons and  $\pi$ -mesons, and perhaps between  $\pi$ -mesons are called strong interactions, and this complex of interacting particles is called the  $\pi$ -nucleon system. Good data are beginning to appear in this field along with new physical concepts, and increasing interest in this system has been evinced by both theorists and experi-

mental groups at Brookhaven. There is a conspicuous "resonance" in  $\pi$ - $p$  scattering at 190 Mev due to the so-called  $T=\frac{3}{2}$ ,  $J=\frac{3}{2}$  state of the  $\pi$ -nucleon system. The effect of this resonance on  $\pi$ -meson production has been studied at Brookhaven and other laboratories, and some of the theoretical predictions have been confirmed by experiment. Many experimental results remain unexplained, however, and recently theorists have proposed the existence of a strong  $\pi$ - $\pi$  interaction between these particles. The suggestion is that these two interactions, the  $\pi$ - $p$  resonances and the postulated  $\pi$ - $\pi$  resonances, may underlie all strong interactions in the 1 to 3-Bev energy range.

The Cosmotron is well suited to this field of investigation, and several experiments are under way. One group, working with hydrogen bubble chamber pictures taken with incident 0.96-Bev  $\pi$ -mesons, has analyzed the process  $\pi^- + p \rightarrow \pi^- + \pi^0 + p$  and the process  $\pi^- + p \rightarrow \pi^- + \pi^+ + n$ . For the first reaction the energy and angular distribution of the final particles gave support to the existence of a strong  $\pi$ - $\pi$  attraction force which held the  $\pi^0$  and  $\pi^-$  together for a short time. One model of the process which is so far acceptable is to picture the incoming  $\pi^-$ -meson as striking a  $\pi^0$ -meson existing in a virtual state near the proton and the two mesons then moving away together, leaving the proton relatively undisturbed. The second reaction, however, does not reveal a corresponding  $\pi^-$ - $\pi^+$  interaction, and more data taken at other incident  $\pi$ -meson energies are needed to determine the accuracy of this simple model.

Proton-proton scattering experiments performed here have also yielded information about the  $T=\frac{3}{2}$ ,  $J=\frac{3}{2}$   $\pi$ - $p$  interaction. Theory predicts that as a result of a  $p$ - $p$  interaction one of the protons can be left in an excited state which subsequently decays in  $10^{-22}$  sec into an ordinary proton and a  $\pi$ -meson by the reaction  $p^* \rightarrow p + \pi$ . With the use of counters the creation of this so-called isobar  $p^*$  was confirmed by observing that scattered protons had an energy and angle appropriate to the formation of the isobar by the process  $p + p \rightarrow p + p^*$ . The existence of the isobar was also confirmed by a group from Yale in an experiment using a hy-

drogen bubble chamber and 2.9-Bev incident protons. Analysis of the interactions  $p+p \rightarrow p+\pi^++n$  showed that the final products occurred through the formation of a doubly charged nucleon  $p+p \rightarrow p^{++}+n$ . These experiments have lent support to the importance of the  $T=\frac{3}{2}, J=\frac{3}{2}$   $\pi$ -nucleon and perhaps to a  $\pi$ - $\pi$  resonance. There remain, however, many unresolved questions.

### Strange Particles

Experiments on the production of characteristics of hyperons and  $K$ -mesons are in progress. In one of these the 14-in. hydrogen bubble chamber is being used to study the interactions of  $K_2^0$ -mesons with protons. It has been possible to obtain a flux of  $\approx 3$   $K$ -mesons per Cosmotron pulse through a liquid hydrogen bubble chamber, and to date  $\approx 3000$  strange particle decays have been observed. So far four  $\Xi^0$  hyperons have been found, which confirms the existence of the  $\Xi^0$  and extends the knowledge of these particles beyond that determined from the one event previously observed at Berkeley.

The 20-in. hydrogen bubble chamber has been exposed to 2.85-Bev protons and 90,000 photographs have been taken. Analysis of these pictures so far has yielded a total cross section for hyperon production of  $0.14 \pm 0.04$  millibarns. This result is significant in view of earlier experiments that gave

conflicting results for the cross section of this fundamental process.

The theory group is working on a number of problems concerned with particle physics. The decay of the  $K$ -meson by the process  $K^+ \rightarrow \pi^+ + \pi^0 + \gamma$  is being analyzed to determine if the resulting gamma-ray spectrum would reveal information about  $\pi$ - $\pi$  interactions. The consequences of  $\pi$ - $\pi$  interactions in the reactions

$$\pi^- + p \rightarrow \begin{cases} \pi^- + p + \pi^0 \\ \pi^- + \pi^+ + N \end{cases}$$

for various isospin states are also being studied, and some previously contradictory results have been explained by the inclusion of a  $T=1$  resonance. Work is also being done on the nature of hyperons which predicts that the magnetic moment of the  $\Lambda^0$  should be  $\approx \frac{1}{2}$  nuclear magneton.

The future program for the Cosmotron is a full one. Several bubble chamber experiments to study the process of associated production near threshold by  $\pi^+$  and  $\pi^-$  mesons are scheduled. In addition, a rather extensive series of hydrogen bubble chamber runs is being planned to obtain further information on  $\pi$ -nucleon scattering around 1 Bev. Counter experiments are now scheduled to measure the magnetic moment of the  $\Lambda^0$  hyperon, to measure production cross sections for  $K$ -mesons and  $\pi$ -mesons from  $p$ - $p$  interactions, and to deter-

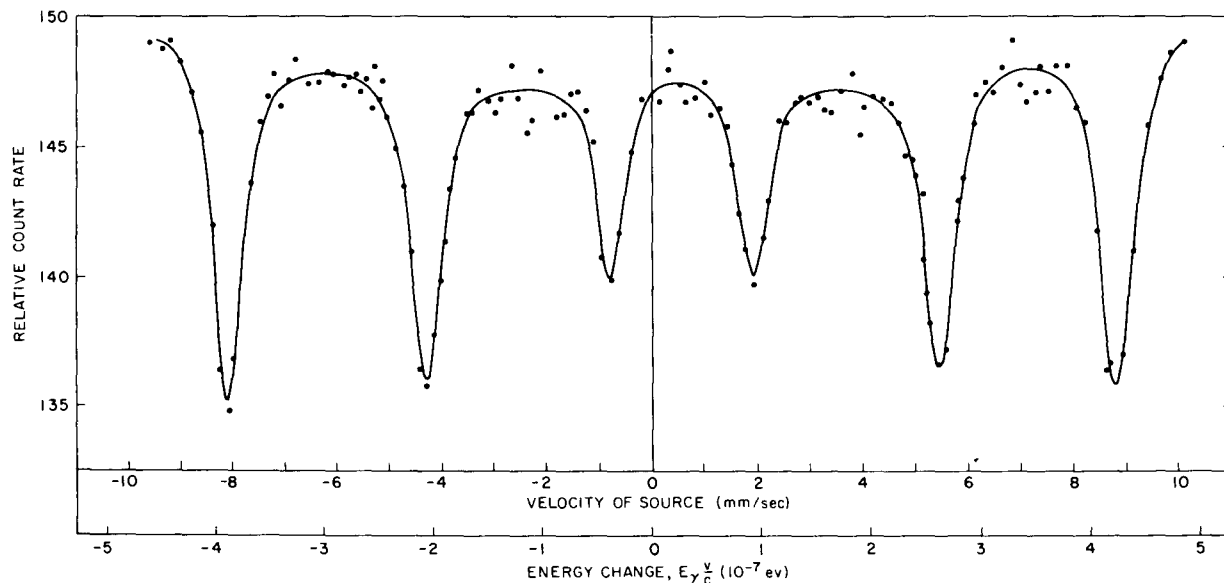


Figure 1. The absorption by  $\text{Fe}^{57}$  bound in  $\text{Fe}_2\text{O}_3$  of the 14.4-kev gamma-ray emitted in the decay of  $\text{Fe}^{57m}$  bound in stainless steel as a function of relative source-absorber velocity. Positive velocity indicates a motion of source toward absorber.

mine the production of  $K_2^0$ -mesons from  $\pi$ - $p$  interactions as a function of incident energy and angle. A run to measure the decay kinematics of the process  $K^+ \rightarrow \pi^0 + \mu^+ + \gamma$  is nearing completion.

Some of these experiments are being performed by experimental groups from universities, others by groups of physicists from universities and from BNL, and some by BNL personnel only. Fully half the experimental effort is being carried out by visiting scientists. In all,  $\approx 106$  physicists are involved, some of them on a part-time basis.

## NUCLEAR STRUCTURE

The researches in this field may be divided into two broad classifications: those dealing with studies of the characteristics of unstable (radioactive) nuclei, and those concerned with the characteristics of the instantaneous products of a nuclear reaction. The term instantaneous, as used here, 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. Studies of the ways in which an unstable nucleus is produced and reverts to a stable nucleus, through the radioactive decay process, yield information on the characteristics of the energy levels of the nuclei involved. Characteristics such as the spin, parity, and lifetime are sought and analyzed to obtain the systematics of the decay process.

Researches dealing with observations of the instantaneous products of a nuclear reaction also yield information about the characteristics of energy levels of nuclei. Some energy levels can be observed both in radioactive decay and in nuclear reaction studies, while others can be observed only in nuclear reactions, because of their very short lifetime. As the techniques in electronic circuits have developed it has become possible to observe lifetimes as short as  $10^{-11}$  sec with reasonably high precision. The ways in which nuclear energy 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 theories of the fundamental structure of atomic nuclei.

The recent discovery of the recoil-free emission and absorption of nuclear gamma-rays in the case of nuclei that are tightly bound in a crystal lattice (the Mössbauer effect) has provided a new technique for the studies of chemical binding effects

and internal magnetic fields in materials. The gamma-ray emitted from a bound nucleus has an extremely small energy spread, and correspondingly the absorption of gamma-rays by such a nucleus shows very sharp resonance. These effects are so marked that a relative motion of only a fraction of a centimeter per second, between a source containing a gamma-emitting nucleus and an absorber containing the corresponding stable nucleus, can change the gamma-ray energy enough to completely eliminate absorption of the gamma-ray by the stable nucleus. The recoil-free emission and resonant absorption of the 14.4-keV nuclear gamma-ray of  $\text{Fe}^{57}$  has been used to determine the quadrupole coupling for the  $\frac{3}{2}$  — excited state of  $\text{Fe}^{57}$  bound in  $\text{Fe}_2\text{O}_3$ , and to measure an energy shift of this nuclear gamma-ray which can be attributed to chemical binding effects. This effect is corollary to the effects of chemical environment on internal conversion coefficients and on electron capture disintegration rates. An energy shift of the order of  $2.5 \times 10^{-8}$  eV is observed. These measurements also yield the value  $5.15 \times 10^5$  oersteds for the internal magnetic field at the position of the  $\text{Fe}^{57}$  nucleus when it is bound in antiferromagnetic  $\text{Fe}_2\text{O}_3$ . Figure 1 shows the data obtained as a function of the relative velocity of the radioactive  $\text{Fe}^{57m}$  in stainless steel and the  $\text{Fe}^{57}$  stable nucleus in  $\text{Fe}_2\text{O}_3$ . Figure 2 shows the energy level schematic for the  $\text{Fe}^{57}$  bound in stainless steel and in  $\text{Fe}_2\text{O}_3$ . The chemical binding effect is the sum of the two energy increments  $\Delta E_1$  and  $\Delta E_2$  shown in this figure.

Theoretical considerations of the recoil-free emission and absorption of gamma-rays have been refined to include the line shift produced by the second-order Doppler effect. It has been found that this shift does not produce any broadening of the Mössbauer line, in agreement with other considerations.

Studies of the radioactive decay of 2.8-hr  $\text{Sr}^{87m}$ , using an internal source technique, show the existence of a  $0.65 \pm 0.25\%$   $K$ -electron capture branch as represented in Figure 3. The low value obtained for  $\log ft = 4.25$  and the small energy release of 115 keV make this  $K$ -branch especially interesting because it establishes  $\text{Rb}^{87}$  as a possible low threshold detector for the inverse neutrino capture reaction,  $\text{Rb}^{87} + \nu \rightarrow \text{Sr}^{87m} + e^-$ , which could be identified by observing the characteristic decay of the  $\text{Sr}^{87m}$  isomer.

A lower limit for the "lifetime" of the electron against decay has been established. Any process in



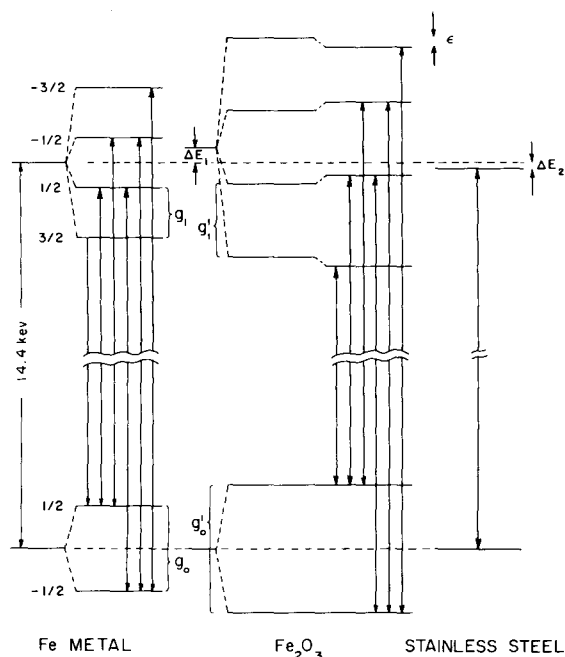


Figure 2. Schematic representation of the ground and 14.4-keV excited states of  $\text{Fe}^{57}$  bound in ordinary iron,  $\text{Fe}_2\text{O}_3$ , and stainless steel. This diagram illustrates the details of magnetic hyperfine splitting, quadrupole interaction, and energy shifts due to chemical binding effects.

which an electron disappears from the  $K$ -shell of an atom, e.g., iodine, can be detected by the subsequent radiation of  $K$  x-rays when the vacancy in the shell is filled by an electron from an outer shell. The amount of iodine  $K$  x-ray present as a background in a large  $\text{NaI}(\text{Tl})$  crystal was used to set a lower limit on any such process. A lower limit of  $10^{18}$  yr was obtained, and this can be considered as a test of charge conservation.

The first case of triple isomerism reported in the literature is that of the  $1.3m$ ,  $21m$ , and  $60d$  isomers of  $\text{Sb}^{124}$ . The large amount of  $L$  x-rays present indicates that the isomeric transitions, although intense, are highly converted. Beta-rays associated with the short-lived isomers have been studied and  $ft$  values determined. On the basis of the beta decay, the known spin and parity of the  $60d$  ground state, and of the single-particle shell model of the nucleus, tentative spins and parities have been proposed for these short-lived isomers. Two gamma-rays not present in the decay of the  $60d$  isomer have been seen in the radiations of the short-lived isomers. Their energies are 485 and 648 keV. Coincidence experiments have shown that these gamma-rays coincide with the 602.6-keV gamma-ray which is the ground-state transition from the

first excited level in  $\text{Te}^{124}$  (see Figure 4). All three gamma-rays have equal intensities. An upper limit of 0.5% was placed on crossover transitions. Angular correlation experiments between the 648 and the 602.6-keV gamma-rays indicated that the 1250-keV level could be assigned a spin 2 and that the 648-keV transition was a mixed  $M1 - E2$  transition with the  $M1$  component predominant ( $E2 \approx 3\%$ ). The 485-keV - 602.6-keV gamma-gamma correlation confirmed these assignments and indicated a spin 0 for the 1735-keV level.

A search has been made for the 4.8-msec isomeric transition in  $\text{Pb}^{205}$ . A transition of 26 keV was established as belonging to the isomeric state and the question of the "missing"  $M4$  transition was tentatively solved.

A collaborative effort to observe the decay of  $\text{O}^{20}$  was carried out with scientists at Los Alamos Scientific Laboratory. The  $\text{O}^{20}$  was produced by bombarding a highly enriched  $\text{O}^{18}$  (95%) gas target with 2.66-MeV tritons. The data yielded a half-life for  $\text{O}^{20}$  of  $13.6 \pm 1.0$  sec and a  $\beta^-$  energy of 2.69 MeV. The proposed decay scheme is shown in Figure 5.

A number of studies have been made of the gamma-rays emitted following the capture of slow neutrons by nuclei. The coincidence spectra of such gamma-rays have been observed for several elements. Studies of normal iron, normal chromium, and the separated isotopes  $\text{Cr}^{50}$  and  $\text{Cr}^{53}$  have essentially been completed and are being prepared for publication.

Work on the nuclear spectroscopy of  $\text{Ir}^{190}$  has established several new, odd-parity levels in  $\text{Os}^{190}$ . Internal conversion coefficients and transition intensities have also been measured and limits have been set for the total disintegration energy of  $\text{Ir}^{190}$ . The results afford an interesting comparison with

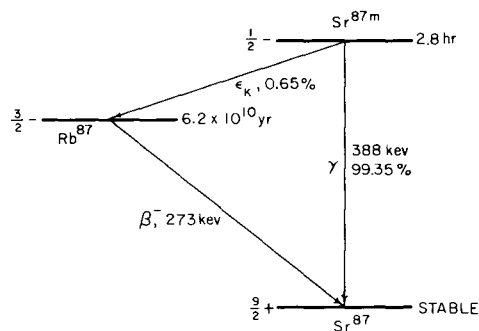


Figure 3.  $\text{Sr}^{87m}$  decay scheme.

the predictions of the asymmetric rotor model of Davydov and Filippov.

A Gerholm electron-electron coincidence magnetic spectrometer, with its auxiliary equipment, has been put into operation. It is expected to be of great use in the continuing program of studies of nuclear structure in the osmium-iridium region, where the nuclear properties are intermediate between those of strongly deformed nuclei and those of spherical nuclei near closed shell. It has already been applied to the study of the decay of  $\text{Ir}^{186}$ .

The energy balance necessary to obtain nuclear resonance fluorescence in  $\text{Cu}^{63}$  is provided by the Doppler shift of the emitted gamma-rays due to the recoil velocity from the beta decay of  $\text{Zn}^{63}$ . Resonance fluorescence has been observed from the 669 and 963-keV resonances with sources in the solid and liquid phases. Since the lifetimes of the emitting states are  $3 \times 10^{-13}$  and  $7 \times 10^{-13}$  sec, respectively, many of the beta decay recoils stop before gamma emission takes place and the resonance yields are much lower than would be observed from a gaseous source. Quantitative measurements of this attenuation have been obtained for both resonances. This motion of copper recoils in a copper lattice has been studied in great detail by Gibson, Goland, Milgram, and Vineyard. By using their model the resonance absorption expected for both copper resonances has been calculated. The agreement between the calculated and measured attenuations is within the error of measurements ( $\approx 10\%$ ). By measuring the attenuation of the resonance and comparing this with the predicted values, it has also been possible to determine uniquely the spin of the 673-keV level in  $\text{Cu}^{63}$  as  $1/2$ . By using a simple model of copper recoil-stopping in water, the resonance attenuation has been calculated to within  $\approx 20\%$  of that observed.

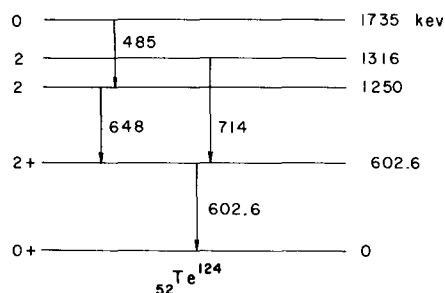


Figure 4. The first four excited levels in  $\text{Te}^{124}$  which are populated in the decay of  $\text{Sb}^{124}$  isomers.

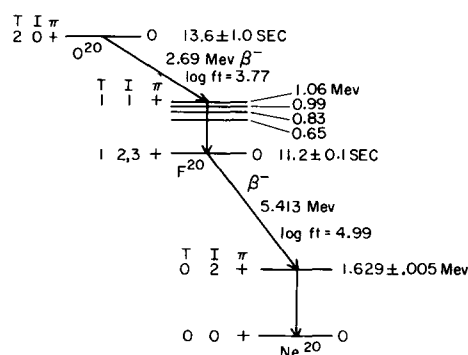


Figure 5. Proposed decay scheme for  $\text{O}^{20}$ . The excited states and the decay scheme of  $\text{F}^{20}$  are from F. Ajzenberg-Selove and T. Lauritsen, *Nuclear Phys.* 11, 1 (1959). The spin and parity assignment of the 1.06-Mev state results from the present work.

The recent development of photomultiplier tubes with small time-jitter has significantly improved the techniques of experimentally measuring the lifetime of an excited state. A conventional 6BN6 time-to-amplitude converter circuit has been used to obtain prompt coincidence time spectra with full width at half maximum of  $5 \times 10^{-10}$  sec and slopes of a factor of two in  $7 \times 10^{-11}$  sec. It has therefore become possible to observe the exponential decay of states with lifetimes as short as  $10^{-10}$  sec. This method is far more accurate and much more easily interpretable than the centroid-shift measurements previously used. The use of a blocking circuit enables the system to accept singles counting rates of  $5 \times 10^4$  counts per sec without loss of time resolution. Careful measurements of the lifetimes of the first excited states of the mirror nuclei  $\text{O}^{17}$  and  $\text{F}^{17}$  have been obtained. The mean lives of these levels are  $\tau$  (0.87-Mev  $\text{O}^{17}$ ) =  $(2.55 \pm 0.13) \times 10^{-10}$  and  $\tau$  (0.50-Mev  $\text{F}^{17}$ ) =  $(4.45 \pm 0.22) \times 10^{-10}$  sec. Since the matrix elements for these transitions are composed of contributions from the  $\text{O}^{16}$  core and the single odd proton or neutron, they are well suited to a careful theoretical study, and surprisingly good agreement with the experimental results has been obtained by R.J. Raz by using a simple collective-model picture.

The lifetime of the 279-keV state in  $\text{Ti}^{203}$  has been carefully measured by W. Deutsch and F. Metzger with the resonance fluorescence technique and use of a high speed rotor. They obtain a lifetime  $\tau_\gamma = (5.14 \pm 0.27) \times 10^{-10}$  sec. This lifetime has now been measured by the coincidence method. The value  $\tau_\gamma = (4.99 \pm 0.10) \times 10^{-10}$  sec is

in excellent agreement with the resonance fluorescence method. This experiment provides the most accurate check of the assumptions entering the interpretation of the resonance fluorescence experiments, and particularly of the theory referring to the Doppler width of resonances in solids.

A compilation of nuclear gamma-rays arranged according to energy has been brought up to date. It has been published as BNL 605 (T-177) and also as part of the 1960 *Nuclear Data Tables* published by the National Academy of Sciences.

An article on the effects of nuclear structure on internal conversion has been completed and will be published in *Ann. Rev. Nuclear Sci.* The formalism developed for the study of nuclear structure effects in internal conversion was successfully applied to the analysis of a number of "anomalous" electric dipole internal conversion coefficients.

A draft was completed of a monograph, *Electromagnetic Radiation From Nuclei*, for the Oxford Press. This is a review of the physical principles involved in gamma-ray emission from nuclei.

The theoretical study of nuclear models and their correlation with the properties of the low-lying nuclear levels continued.

An experiment is being conducted under the Columbia-BNL cooperative program to measure the beta-neutrino angular correlation coefficient in the decay of  $\text{He}^6$  by a new method. Calculations have shown that the shapes of the energy spectra of recoil nuclei which have emitted beta-particles of selected energy are more sensitive to the value of this angular correlation coefficient than the measured variables in previous experiments on  $\text{He}^6$ . The experimental apparatus and generator to produce the  $\text{He}^6$  gas according to the reaction  $\text{Be}^9(n, \alpha)\text{He}^6$  have been constructed and installed at the BNL Graphite Research Reactor. The energy of the beta-particles is measured with a plastic scintillation spectrometer of the well type, and the energy of the corresponding recoil nuclei is obtained with the aid of an electrostatic analyzer and time-of-flight system. The shape of the beta-energy spectrum, the half-life of the decay, and the spin of the  $\text{He}^6$  nucleus have been determined preliminary to the final measurements.

The charged particles available from the Van de Graaff accelerator were used for a number of problems relating to the structure of light nuclei. Much of the effort was concerned with the energy levels in  $\text{C}^{12}$ , particularly the 7.66-Mev second excited state which is thought to be involved in the

burning of helium in red giant stars. Although this level was known to be unstable against alpha-particle emission, experimental proof that carbon could be formed from helium was lacking, since neither the transition from the 7.66-Mev level to the first excited state nor that to the ground state had been observed. After preliminary experiments at BNL had indicated that the ground-state transition might be detected by using the intermediate-image pair spectrometer together with the  $\text{Be}^9(\alpha, n)\text{C}^{12}$  reaction, the spectrometer was moved to Oak Ridge National Laboratory for 5 wk during the summer of 1959 in order to carry out the measurements. This move was necessary because of the higher energy alpha-particle beam available from the Oak Ridge Van de Graaff accelerator. The 7.66-Mev ground-state transition was observed with an intensity of 1/200,000 relative to the alpha-particle decay rate.

A search for the 3.2-Mev transition from the 7.66-Mev level to the first excited state of  $\text{C}^{12}$  was made at the Brookhaven Van de Graaff accelerator by means of a triple coincidence measurement on the  $\text{B}^{10}(\text{He}^3, p)\text{C}^{12}$  reaction. Instead of finding the transition in question, the experiments revealed the presence of a new energy level in  $\text{C}^{12}$  at 9.0 Mev. A further search for the missing 3.2-Mev transition is being made through the beta decay of  $\text{B}^{12}$  to  $\text{C}^{12}$ .

The pulsed beam facility of the Van de Graaff accelerator has been used in studies of the lifetime of the 0.87-Mev first excited state in  $\text{O}^{17}$ . Since this lifetime is short compared with the pulse duration obtainable with this facility, it was not possible to make precise measurements of the lifetime of this excited state. By means of new ultrafast coincidence techniques, however, it has been possible to measure the lifetimes of the first excited states in  $\text{O}^{17}$  and in  $\text{F}^{17}$ . These lifetimes are both in the  $10^{-10}$ -sec range.

## 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 Graphite Research Reactor, investigations have been made of the scattering of very slow or "cold" neutrons by liquids and solids.

The diffusive motions of liquids can be studied by measuring the small energy changes that occur

when cold neutrons are scattered from liquid samples. These measurements can be related to the self-diffusion of liquid molecules by means of a theoretical model suggested by G.H. Vineyard of Brookhaven, based on a general theory of the scattering of neutrons by liquids first proposed by L. Van Hove. A series of measurements in liquids  $\text{H}_2\text{O}$ , lead, and tin have been performed during the last year to study these related effects.

In the experimental arrangement a beam of slow neutrons is obtained by filtering the neutrons from the BNL Graphite Research Reactor through beryllium. These slow neutrons are then incident on the various liquid samples, and the analysis of the energy spectrum of the scattered neutrons is carried out at  $90^\circ$  to the incident beam by using the Brookhaven slow chopper equipment. Measurements were carried out at several temperatures for each sample. The experimental arrangement yields a measurement of the correlated motions of atoms separated by approximately the nearest-neighbor distance in the liquids and is sensitive to time intervals of the order of  $10^{-13}$  sec. The results for  $\text{H}_2\text{O}$  give no measurable diffusive broadening. These results can be understood in terms of a model for water in which the  $\text{H}_2\text{O}$  molecule executes a number of vibrations about some average equilibrium position before making a diffusive transition (jump). In lead and tin experimental evidence of diffusive broadening is observed. However, the magnitude of the broadening is less than expected from tracer and nuclear resonance diffusion measurements, which indicates that the classical ideas of atomic motions in liquids may be in error.

The BNL fast chopper with its time-of-flight analyzer has been used mainly in two areas of interest in the field of neutron spectroscopy. The first of these is the remeasurement of total radiation widths that have been reported to fluctuate widely from level to level in the same nuclide. Since total radiation widths are expected to be very nearly the same for levels in the same nuclide, it was of considerable interest to re-examine cases found to deviate from this expectation. Levels in antimony and mercury were investigated in detail. In  $\text{Sb}^{121}$ , the 6.23-ev level was previously reported to have a radiation width approximately one half as large as those of the 15.4 and 29.7-ev levels. This investigation yielded widths of  $90 \pm 4$ ,  $88 \pm 10$ , and  $89 \pm 10$  mv, respectively, in agreement with previous expectations.

In mercury, several pairs of levels have been investigated, with a pair in each of three isotopes. For  $\text{Hg}^{198}$  levels at 23 and 91 ev, radiation widths of  $130 \pm 14$  and  $174 \pm 23$  mv were obtained. In the case of  $\text{Hg}^{199}$  the present work gives a value of  $376 \pm 95$  mv for the 131-ev level, to be compared with  $310 \pm 40$  mv for the 33-ev level as quoted by Levin and Hughes. In contrast to these results, in  $\text{Hg}^{201}$ , for levels at 43.3 and 71.8 ev, the present work shows values of  $241 \pm 36$  and  $470 \pm 70$  mv. This difference is outside experimental error.

The second area of interest in which the BNL fast chopper has been used is a continuing study of the total cross section of nuclides with atomic weights in the range  $\approx 100$  to 120, in which the reported *s*-wave neutron strength function measurements show the greatest divergence from theoretical optical model predictions. The cases of rhodium, tellurium, and ruthenium have been investigated. In each case consistent strength function determinations for each element have been obtained, both from the measured level parameters of individual levels and from the average cross section in the region from  $\approx 2$  or 3 kev down to  $\approx 500$  ev. The strength function values obtained for rhodium, tellurium, and ruthenium are  $0.6 \pm 0.1$ ,  $0.6 \pm 0.2$ , and  $0.3 \pm 0.2 \times 10^{-4}$ , respectively. These values are in agreement with the experimental values and with the trend exhibited by neighboring nuclides and show the characteristic disagreement with the theoretical calculation of Weisskopf and Feshbach in this region of atomic weight.

For these same nuclides the neutron widths and energies of individual levels have been obtained in the energy region from  $\approx 10$  to 400 ev above the neutron binding energy. Isotopic assignments for almost all levels in tellurium and ruthenium have been determined. In the case of ruthenium, determinations of total widths from 9 of the levels have been made, and for 5 of these the total radiation widths have also been obtained.

The joint BNL-Chalk River fast chopper program is continuing. A 90-meter flight path and detector station has been completed by Atomic Energy of Canada Limited, and a bank of 380  $\text{BF}_3$  counters supplied by BNL has been installed in the station. This installation is intended for use in high resolution neutron transmission measurements of the type carried out at Brookhaven during the past five years to be extended to higher neutron energies (100 ev to 10 kev). An over-all resolution of  $\approx 15$   $\mu\text{sec}/\text{meter}$  has been checked

experimentally by examining resonances in manganese and  $U^{238}$ . Under these conditions a net counting rate of  $\approx 1$  count per min per  $\frac{1}{2}$ - $\mu$ sec channel is obtained. This is comparable to the counting rate obtained at Brookhaven with the shorter 20-meter flight path. Total cross section measurements of vanadium, zirconium, and molybdenum are now being carried out by the group at Chalk River.

The study of transition probabilities of gamma-rays emitted from various discrete energy levels in the compound nucleus is being continued by the BNL group at Chalk River with the gamma-ray detection apparatus originally assembled at Brookhaven. It was expected that the transition probabilities for resonance levels of the same spin and parity would fluctuate widely from level to level corresponding to a single-channel process for radiation to the same final state. However, the first measurements of the ground-state transitions in  $W^{184}$  revealed much smaller fluctuations than expected for a one-channel distribution. Measurements have now been extended to platinum, mercury, and  $U^{238}$ .

In  $Pt^{195} + \text{neutron}$ , transitions to the ground state of  $Pt^{196}$  have been observed from 10 resonances between 11 and 300 ev. The distribution of relative transition probabilities was found to fit a  $\chi^2$  distribution with  $\approx 5$  degrees of freedom. A single-channel process according to the theory of Porter and Thomas should correspond to a  $\chi^2$  distribution with one degree of freedom.

In the case of mercury, the transition probabilities show more fluctuation. However, isotopic assignments are not known with certainty for many of the levels measured, and therefore the interpretation of the transition probabilities required clarification of these questions.

For tungsten, platinum, and mercury some small ambiguity in the interpretation of the results exists because of the possibility of incorrect spin assignment of the resonance state. In order to avoid this problem, an even  $A$  - even  $Z$  target nucleus ( $U^{238}$ ) was chosen for investigation. These measurements are now nearly completed. The thermal neutron capture gamma-ray spectrum for  $U^{238}$  is dominated by a single line at 4.06 Mev. This line was chosen for the resonance capture studies. Ten levels were observed between 6 and 211 ev, and the computed transition probabilities show little fluctuation from level to level. Calculations for these levels show a fit to a  $\chi^2$  distribution

with  $\approx 50$  degrees of freedom. These data are strong evidence that the single-channel analogy between the decay of the compound nucleus by the emission of a neutron and the decay by the emission of a specific gamma-ray is not valid.

The accumulation of new data on neutron cross sections and the refinement of old measurements made it desirable to prepare a supplement to BNL 325, *Neutron Cross Sections. Supplement No. 1* to the second edition of BNL 325 has been published. It contains 129 pages of tables, curves, and references representing the accumulation of new data during the period from July 1958 through January 1960. A comparison of the data in the *Supplement* with the second edition of the compilation shows that the bulk of new data is in the field of capture cross sections. Better experimental data are also available for  $\alpha$ , the ratio of capture to fission cross section for the fissionable materials. It is evident that one very useful function of the compilation is pointing out the need for new and better data.

Crystal spectrometers continue to be used at the BNL Graphite Research 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. Some routine measurements of neutron cross sections have been continued, with most of the effort being directed toward the precise measurement of isotopic cross sections for the rare earths in the energy region 0.01 to 1.0 ev. These data are of current interest in reactor physics. Data were completed on  $Er^{167}$ ,  $Gd^{155}$ ,  $Gd^{157}$ ,  $Lu^{176}$ ,  $Eu^{151}$ ,  $Dy^{164}$ , and  $Sm^{152}$  (6 to 10 ev).

Under the cooperative program between Columbia University and BNL, studies of nuclear and molecular phenomena are being conducted with a single-crystal neutron spectrometer at the BNL Graphite Research Reactor. Techniques developed under a continuing program of spectrometer improvement have made available monoenergetic neutron beams of any desired energy between 0.0006 and 10 ev. Emphasis has been placed on precision cross section measurements at thermal and subthermal neutron energies. The absolute values of the total neutron cross sections of  $U^{233}$  and  $^{235}$  were measured at neutron energies between 0.000818 and 0.0818 ev. Previous data measured in this region have shown discrepancies of 3 to 4%. More accurate cross section data were needed for reactor theory and design. In these measurements two types of samples, metallic foils

and D<sub>2</sub>O solutions of uranium nitrate, were measured for each uranium isotope. The measurements with the metallic foils yielded  $\sigma_t(\text{U}^{233}) = 586 \pm 2$  b and  $\sigma_t(\text{U}^{235}) = 698.7 \pm 4.7$  b at the standard thermal energy of 0.0253 ev. The measurements with the liquid uranium nitrate solutions yielded  $\sigma_t(\text{U}^{233}) = 587 \pm 5$  b and  $\sigma_t(\text{U}^{235}) = 695.0 \pm 1.8$  b at 0.0253 ev. In both cases, the foil measurements agreed with the liquid sample measurements to a precision of better than 1%. The resulting value of  $\sigma_t(\text{U}^{235})$  at 0.0253 ev, when combined with the previously measured value for  $\text{U}^{235}$  of  $(1 + \alpha) = \sigma_a/\sigma_f$  at 0.0253 ev, yields a value for the  $\text{U}^{235}$  fission cross section at 0.0253 ev of  $\sigma_f(\text{U}^{235}) = 590.8 \pm 5.4$  b.

The total neutron cross section of  $\text{B}^{10}$  has been measured by the Columbia group at neutron energies between 0.00291 ev and 0.1 ev with the crystal spectrometer. The neutron absorption cross section of  $\text{B}^{10}$  has been obtained in the past from the measured neutron cross section of natural boron together with a mass spectrometer determination of the  $\text{B}^{10}$  content of the sample. The values of the neutron absorption cross section of natural boron are in good agreement ( $\pm 0.7\%$ ). However, mass spectrometer measurements of the  $\text{B}^{10}$  content of natural boron vary between 18.4 and 20.0 atom %  $\text{B}^{10}$  and give rise to a variation of  $\pm 4\%$  in the calculated  $\text{B}^{10}$  cross section. The availability of highly enriched  $\text{B}^{10}$  samples, the improved reliability of mass spectrometric determinations of the boron isotopic abundance, and the high degree of precision attained for neutron transmission measurements in the thermal energy region have made possible a direct and precise measurement of the  $\text{B}^{10}$  cross section. The need for reducing the uncertainties on the boron cross section arises from its widespread use as a standard for measurement of other cross sections and from its use in boron loading specifications in reactors. The  $\text{B}^{10}$  absorption cross section was obtained by subtracting the relatively small known scattering cross section from the measured total cross section. Two samples enriched to  $92.84 \pm 0.06$  atom % and  $99.88 \pm 0.01$  atom %  $\text{B}^{10}$  were used for these measurements. A sample of Argonne-Brookhaven standard normal boron,  $19.8 \pm 0.1\%$   $\text{B}^{10}$ , was measured simultaneously. The resulting absorption cross section for normal boron was  $\sigma_a(0.0253 \text{ ev}) = 764 \pm 3$  b and obeyed the relation  $E^{1/2}\sigma_a = 121.5 \pm 0.5 \text{ b(ev)}^{1/2}$ . The average of the results for the two enriched samples yields  $\sigma_a(\text{B}^{10}) = 3838 \pm 11$  b at 0.0253 ev

and obeys the relation  $E^{1/2}\sigma_a(\text{B}^{10}) = 610.5 \pm 1.7 \text{ b(ev)}^{1/2}$ . The ratio of the above values of the absorption cross sections for  $\text{B}^{10}$  and normal boron yields a value of  $19.9 \pm 0.1$  atom %  $\text{B}^{10}$  for the normal boron sample, which is in good agreement with mass spectrometer determinations.

In cooperation with the Chemistry Department of Columbia University, a study is being made of slow neutron scattering by hydrogenous solids, liquids, and gases. Transmission measurements have been completed on four ammonium halide salts,  $\text{KH}_2\text{PO}_4$ , four liquid methylbenzene compounds, and  $\text{NH}_3$  gas. These measurements have covered a neutron energy range from 0.0006 to 2 ev. The cross sections per molecule calculated from the transmission data have been corrected for neutron absorption and divided by the number of hydrogen atoms per molecule to obtain the total scattering cross sections per proton for each of these compounds. When the cross section data are plotted as a function of neutron wavelength in the subthermal neutron energy region, a definite correlation is observed between the slopes of the curves and the proton dynamics and the strength of chemical binding in these compounds. Theoretical calculations of the cross sections for the gas and liquid samples, based on the Krieger-Nelkin theory, agree well with the observed data. An apparatus for maintaining samples at low temperatures has been constructed for use with the Columbia University neutron spectrometer. Temperatures between  $+20^\circ$  and  $-196^\circ \text{C}$  can be obtained and held to within one degree. With this apparatus, preliminary total cross section measurements have been made on  $\text{MnO}$  through the available temperature range at several neutron wavelengths in order to study the variation of inelastic scattering with temperature. Curves of the total cross section of  $\text{MnO}$  and  $\text{MnF}_2$  as a function of neutron energy were also taken at room temperature and at  $-196^\circ \text{C}$ .

A program is under way to use polarized nuclei as targets for polarized monochromatic neutrons in order to determine the  $g$ -value for various slow neutron resonances. Efforts have continued on the development of cryogenic techniques which will be adequate to produce appreciable nuclear polarization. Temperatures of the order of  $0.01^\circ \text{K}$  have been produced by two stages of adiabatic demagnetization. Some systems tested have remained below  $0.02^\circ \text{K}$  for periods of 1 hr or more. Heat leaks as low as 25 ergs/min have been observed at



0.01° K which are accounted for by leakage through the lead superconducting switch used. Several useful techniques have been devised; e.g., it has been demonstrated that graphite has several useful properties at ultralow temperatures which make it particularly suitable for fabricating mechanical supports for the cold assembly. A new type of thermal valve was developed which promises to improve the low temperature duty cycle by a large factor. Techniques for preparing the paramagnetic salts appear to yield superior heat transfer characteristics at the low temperatures. An iron core magnet, cooled by liquid nitrogen, has been tested which provides a magnetic field of 15 kilogauss over a gap of 1½ in. using only 1 kw of power.

The polarized neutron beam has been tested and polarizations of >98% are obtained from the (111) and (200) Bragg reflections from a cubic crystal of 94% Co - 6% Fe alloy. The direction of polarization can be rotated by various guide field arrangements without detectable loss of polarization. In addition, the magnetic guide field can be reversed with respect to the polarization direction with only 3 to 4% loss of polarization. The polarization spectrometer and target cryostat are now completed and almost fully assembled. Final testing and adjusting are in progress.

A program is under way at the 18-in. cyclotron to study the scattering of polarized neutrons in the energy range 300 kev to 2 Mev. The polarized neutrons are produced by the (*p,n*) reaction in lithium. Techniques have been developed to obtain efficient neutron detection down to energies of 300 kev or less by effective discrimination against gamma-ray response in a scintillation detector. The neutrons of different energies are separated in the detection equipment by utilizing the time-of-flight technique.

#### ATOMIC AND MOLECULAR PHYSICS

The techniques of atomic beams and paramagnetic (electron) resonance are being used in a continuing program of measurements of nuclear spins and magnetic moments.

Work is under way to measure nuclear spins and related properties of radioactive nuclei by using principally an atomic beam resonance apparatus. The principal effort during the past fiscal year has been directed to a precision measurement of the nuclear magnetic moment of  $\text{Cs}^{134m}$  (3.1 hr).

The purpose of this experiment is to determine the hyperfine-structure anomaly. Preliminary data indicate that this nucleus has an anomaly considerably larger than any heretofore observed, of the order of a few percent. Work is being done to refine the measurement. A new measurement of the hyperfine-structure interactions has been made of the electronic ground state of  $\text{Cs}^{134m}$  yielding the value  $3684.5780 \pm 0.0015$  Mc/sec.

Some exploratory work has been done on a method of introducing a paramagnetic vanadium ion into a MgO crystal in an attempt to determine the spin of  $\text{V}^{49}$ . Such a process may be more fruitful than the aqueous solution previously used without success.

In cooperation with the BNL Chemistry Department and the University of Wisconsin a sample of  $\text{Ar}^{39}$  has been prepared and is being studied for a determination of the nuclear spin by optical interference spectroscopy.

Guest scientists from Princeton University have continued their investigation of the magnetic moment of  $\text{Na}^{21}$  as part of a program of measurements of mirror image nuclei. The  $\text{Na}^{21}$  nuclei are produced from neon by charged particles accelerated in the BNL 60-in. cyclotron. Some success has been achieved in aligning the sodium by optical pumping.

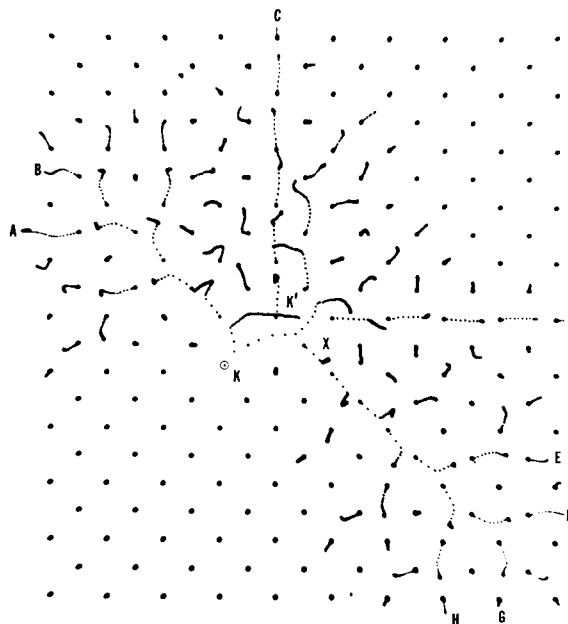


Figure 6. Orbits in a cubic plane of copper produced by a 400-ev knock-on atom starting at *K*, directed 10° away from the [011] axis. Knock-on has gone to *K'*, and collision chains *A*, *B*, ... *H* have reached the boundaries.

## SOLID STATE PHYSICS

## Theory

Most of the theoretical work during the year has been on the production and properties of defects in crystals. Several calculations have reached the stage of giving important results.

Machine calculations of radiation damage dynamics have continued with accumulation of excellent data. The computational procedures have been greatly improved, and a successful scheme has been developed whereby the results of the computations may be presented as a moving picture of the various time sequences in damage events and processes. Early results on low energy damage events, described last year, already indicated the power of this technique for studying the intricate atomic motions associated with the interaction of high energy radiation with matter. Low and moderate-energy damage events and the properties of a number of simple point defects in copper are now well understood. The interstitial has been shown to possess only tetragonal symmetry, instead of full cubic symmetry as was generally supposed. The crowdion has been shown to be unstable, but a dynamic crowdion has been shown to propagate along both  $\langle 110 \rangle$  and  $\langle 100 \rangle$  directions. Various Frenkel pairs (interstitial-

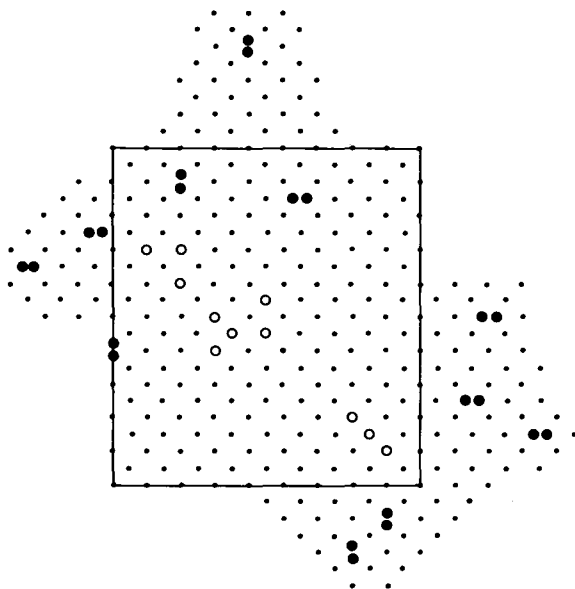


Figure 7. Estimated array of 11 vacancies (open circles) and 11 tetragonal interstitials (double dots) produced by the event of Figure 6. The original set of atoms is outlined by the rectangle. Thirty-nine replacements have occurred. Some of the vacancy clusters may spontaneously rearrange.

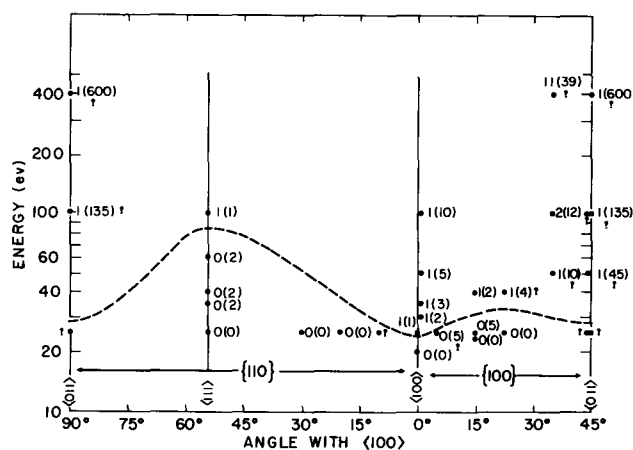


Figure 8. Diagram showing energy and angle of knock-on in all radiation damage events calculated (for copper) in which knock-on lay either in  $\{100\}$  or  $\{110\}$ . The first figure in each case is number of displacements (Frenkel pairs); the figure in parentheses is number of replacements. Dotted line shows threshold energy for displacement.

vacancy pair) have been examined, and the limiting separation for stability in various directions has been determined. Pairs oriented along the  $\langle 110 \rangle$  direction must be separated to the fourth-neighbor position in order to be stable.

A few typical examples of dynamic radiation events are shown in the following figures. Moving picture sequences have been run in sets containing up to 998 atoms ( $2 \times 9 \times 10$  unit cells), representing copper. One of the pictures is shown in Figure 6. Orbits in a cubic plane of atoms are shown. The knock-on was at  $K$  and was endowed with 400 eV, its velocity vector lying  $10^\circ$  away from the  $[011]$  direction. At the end of the run the knock-on has moved to  $K'$  and collision chains  $A, B, \dots H$  are still active. From what is known about these collision chains, the final configuration can be estimated, and involves the set of interstitials (double dots) and vacancies (open circles) shown diagrammatically in Figure 7. It is possible that some of the close-lying vacancies here will rearrange into other clusters. The tendency for interstitials to be outside the vacancies and for trivacancies and higher clusters to be produced is evident. A summary of all dynamic events in which the knock-on velocity lay either in  $\{100\}$  or  $\{110\}$  is given in Figure 8. Each run is represented by a dot in a plane showing knock-on kinetic energy and direction. The first figure appended to each dot gives the number of Frenkel pairs produced, and the

figure in parentheses gives the number of replacements. The dotted line shows the present best estimate of the threshold energy at each direction for producing at least one Frenkel pair.

A closely related topic is the annealing of radiation-induced or quenched-in defects. Recent theoretical work has shown that impurities, even when present at very low concentrations, can seriously influence the annealing kinetics. The general equations for the annealing of vacancies in metals containing impurities to which the vacancies can be attached have been solved on an analogue computer for a wide variety of parameters. The computer results show that some simplifying assumptions may be made which permit the general equations to be solved analytically. It is shown that for many physically interesting cases of vacancy migration the decay is exponential, and the decay constant is related to, but not equal to, the rate constant for vacancy migration. It is further shown that only experiments performed on zone-refined metals can give the correct vacancy migration energy, and that impurity contents as low as  $10^{-5}$  can seriously affect the results. The calculations indicate that careful annealing experiments on impure metals prepared by controlled doping can be used to measure the binding energy of vacancies to impurities.

The theory of the kinetics of color center formation in insulators has been developed further. It has been extended to include the case of many color centers with certain restrictions, namely those of very rapid or very slow annealing. This treatment has been applied to the coloring of a particular glass sample that contains four different color centers. The theoretical expressions describe adequately the growth of gamma-ray induced coloring in this glass.

The nature of the interaction potentials for various substances is of crucial importance in many of the above calculations. Some advances have been made in this field. Corrections to the Thomas-Fermi model for a many-electron system have been developed. A method has been devised for breaking up the electron distribution function of the Thomas-Fermi atom into components corresponding to the angular quantum number. The characteristics of the effective electron shells obtained in this way agree satisfactorily with those obtained from wave mechanics in the following respects: (a) median radius of  $K$ ,  $L$ ,  $M$ , ... shells; (b) contributions to the diamagnetic susceptibility

of the inert gases; and (c) coherent and incoherent scattering of x-rays by the inert gases. A generalization of a statistical procedure for calculating the forces between pairs of atoms has also been accomplished, resulting in improved pairwise interaction potentials for various substances.

Some further work has been done on the theory of liquids. A theoretical demonstration has been given that the law of corresponding states can be extended to predict correspondences in inelastic neutron scattering from fluids. A discussion of general molecular distribution functions involving two times has also been completed.

Some work has also been done on the use of nuclear resonance radiation in the study of solids. In particular, a theoretical study has been made to determine the feasibility of using nuclear resonance radiation to investigate the change in the lattice vibrational spectra upon the introduction of impurities capable of absorbing or scattering nuclear resonance radiation. Preliminary results indicate that such measurements of the perturbed vibrational spectra are possible and will also yield information on the anharmonic terms in the series expansion of the crystal potential.

### Radiation Effects

Radiation effects and other departures from perfect periodicity are under investigation with many diverse techniques. This is one of the major experimental research activities in solid state physics.

There has been considerable speculation about the production and properties of thermal spikes during irradiation, but no clear-cut experimental evidence has been found for or against the existence of such spikes except in fissionable materials. Some preliminary work on the gray-to-white tin transformation was described last year which indicated that thermal spikes were of insufficient intensity to produce any gray-to-white transformation. This experiment has been greatly refined and carried out at a temperature, namely  $0^{\circ}\text{C}$ , at which any reversion of white-to-gray tin is minimized. The results are the same as before; no significant change was found in the amount of white tin present as a result of irradiation. According to the conventional theory of thermal spikes, 16% of the gray tin should have been transformed to white during this particular irradiation. The experimental results indicate strongly, therefore, that the theory of thermal spikes needs considerable revision.

Other solid state reactions, controlled by nucleation and/or diffusion, are under study. The study of radiation-enhanced diffusion is being continued. An investigation of the flux dependence of radiation-enhanced ordering in  $\alpha$ -brass has been completed recently, in cooperation with Los Alamos, with a nuclear rocket engine used as a source of fast neutrons. Identically prepared brass wires were simultaneously irradiated at 60°C at different distances from the engine, and thereby in different fluxes. Electrical resistivity changes, reflecting short-range ordering by enhanced diffusion, were measured. Diffusion constants calculated from the resistivity relaxation times were essentially linear in the flux, in agreement with the simple theory. Enhancement of diffusion by gamma-rays has also been observed, and an extensive set of data is being accumulated at various temperatures and fluxes. The data now available show that the gamma-ray enhanced diffusion coefficient depends on the temperature. This indicates a different annealing mechanism from that during fast neutron irradiation, probably because interstitial-vacancy recombinations are favored. The existence of enhanced diffusion was also demonstrated visually in an experiment completed recently. Helium precipitation in copper, a process that depends on the diffusion of vacancies to provide space for the helium coagulation, was found to be enhanced by reactor irradiation during the required annealing treatment at 435°C. Photomicrographs showed far more copious void formation in the irradiated than in the unirradiated samples. Electron microscope studies showed that annealing in the reactor increased the size of the voids formed.

Other alloy systems, representative of enhanced diffusion or enhanced nucleation, are being investigated. Typical examples are the copper-nickel (a segregating alloy) and the gold-platinum (a precipitating alloy) systems. Preliminary results have been obtained on the precipitation of carbon in iron. Somewhat unexpectedly, neutron irradiation was found to enhance the carbon precipitation as measured by internal friction techniques. The characteristic time constant for the precipitation has been decreased by as much as a factor of a thousand by reactor irradiation. A preliminary analysis of the data indicates that irradiation primarily increases the number of nuclei for the carbon precipitation reaction.

The work on radiation effects in diamond, a purely covalent crystal, continues. Diamond sam-

ples have now been irradiated in the Materials Testing Reactor up to an integrated fast flux of  $13.7 \times 10^{20}$ . The density of these irradiated diamonds was found to be 1.96 g/cm<sup>3</sup>, independent of physical size (from  $6 \times 10^{-9}$  to  $5 \times 10^{-3}$  cm<sup>3</sup>); i.e., the density is less than that of graphite (2.25 g/cm<sup>3</sup>) and represents a decrease of 44.3% from that of unirradiated diamond. The relation between density change and irradiation dose remains linear over this large range of exposure. This suggests that the change in density of diamond can be used as a rather precise fast flux dosimeter for irradiations carried out at temperatures of 100°C or less. After such heavy irradiations the diamond has a glasslike or amorphous structure but retains its external crystalline features (see Figure 9). The hardness is approximately that of the hardest nitrated steels (Vickers hardness 1150). Because of its amorphous structure its physical properties, such as hardness and x-ray pattern, are independent of orientation, as shown in Figure 9.

It is surprising that such a heavily irradiated and badly distorted solid holds together at all. It is even more surprising that this heavily irradiated diamond is very stable. Annealing studies show that for temperatures of 1000°C or more a further decrease in density of 5% occurs, while for lightly irradiated diamonds annealing produces an increase in density or recovery to diamond. Final interpretation must await further work, but it is clear that this is a very unusual form of solid carbon.

Optical measurements on alkali halides and other insulators are yielding important basic information about the number and character of radiation-induced defects. Coloring measurements on reactor and gamma-ray irradiated alkali halides have become reproducible and can now be used to determine the concentration of defects produced by reactor irradiation. Curves of *F*-center coloring vs gamma-ray dose rise rapidly below  $10^6$  r and then level off to a constant slope. If a gamma-ray coloring curve is interrupted with a short reactor irradiation, a stepwise increase in the curve is introduced. In this way it is found that reactor irradiation markedly increases *F*, *V*, *M*, *R*, and other bands, and the absorption from 3 to 6 ev. The slope before and after short (30-min) reactor irradiations is the same. From the stepwise increase the number of vacancies introduced by reactor irradiations can be computed. Quite good agreement is found with the theories of Kinchin and Pease and Seitz and Koehler.

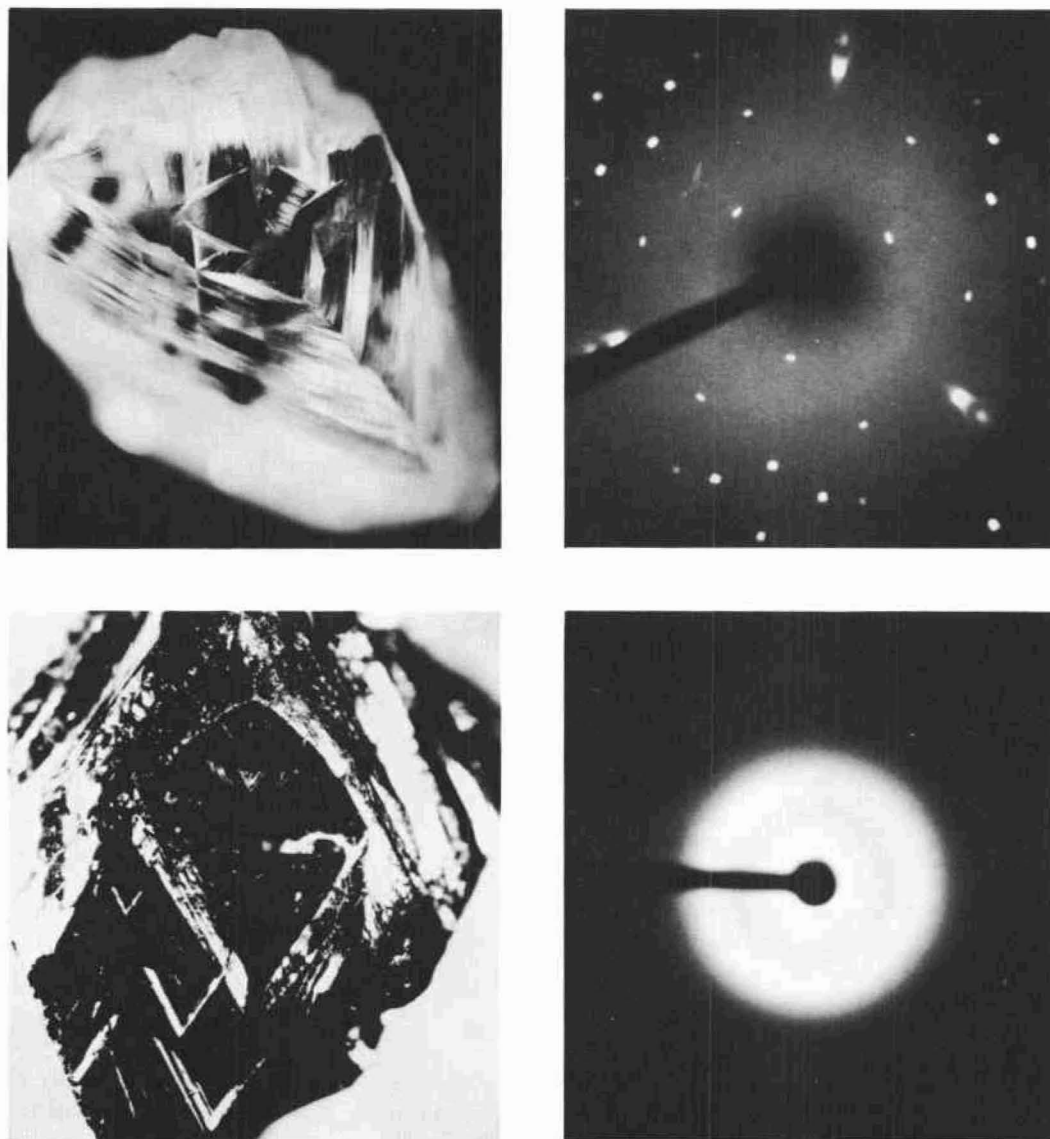


Figure 9. Top: Unirradiated diamond, exhibiting growth trigon; and transmission Laue taken normal to picture (x-ray beam parallel to  $[111]$ ), exhibiting characteristic threefold symmetry. Bottom: Diamond irradiated to  $13.7 \times 10^{20}$  nvt (fast), and transmission Laue taken with identical exposure. Although the irradiated diamond retains its external crystalline features, its internal structure is amorphous.

A careful determination of the annealing characteristics of the reactor-induced color centers in fused silica is very close to completion. Below  $270^\circ\text{C}$  all the coloring removed by the anneal can be restored by subsequent gamma irradiation, while only a fraction of the color removed above this temperature is restored. This indicates clearly that all the color removed below  $270^\circ\text{C}$  results from an electronic process, while above this temperature the reactor irradiation-induced defects responsible for the color centers are annealed out.

Similar annealing experiments are under way on irradiated  $\text{Al}_2\text{O}_3$ . The separate removal of trapped electrons and crystal defects is also observed in this material, but the separation is not so clear cut as in the case of silica.

The cooperative program with Brown University, now in its sixth year, is concerned primarily with the application of ultrasonic techniques to the study of irradiation-induced changes in solids. Equipment has been developed for in-pile ultrasonic velocity and attenuation measurements. A

successful in-pile experiment has been run on one  $\alpha$ -quartz sample. Reactor irradiation caused an increase in attenuation which was still rising after it had reached a value twice that of the unirradiated material. Pre- and postirradiation measurements in  $\alpha$ -quartz showed that irradiation causes a drastic change in the attenuation vs frequency characteristics of this crystal. Attenuation has also been measured as a function of temperature; these curves show that there is a large amount of annealing near 350°C. Another subject of investigation has been a low temperature study of gamma-ray induced changes in ultrasonic attenuation in sodium chloride. The results obtained at liquid nitrogen temperature show that dislocation pinning proceeds at such a rapid rate that thermally activated processes cannot be controlling in producing these effects in sodium chloride. A possible interpretation in terms of the interaction of charged defects with charged dislocations is being investigated.

The cooperative program with Picatinny Arsenal has been continued. A series of optical absorption measurements at room and liquid nitrogen temperatures have been made on gamma-ray colored  $\text{KN}_3$ . Two results emerge. First, lowering the temperature shifts the high energy (ultraviolet) absorption edge, and second, there is surprisingly little narrowing of all the absorption bands upon cooling. This indicates that the defects associated with the bands are only slightly coupled to the lattice. An interesting electron spin resonance spectrum has been found (in cooperation with the University of Connecticut) in  $\text{KN}_3$  at liquid nitrogen temperature after irradiation with ultraviolet light or gamma-rays. The hyperfine structure and the orientation dependence of the spectra indicate that the radiation-induced defect is a linear  $\text{N}_4^-$  molecule-ion.

The effect of radiation on the chemical activity of various solids is being investigated. A study of  $\text{KBrO}_3$  gave some unusual results. Decomposition rate vs temperature plots show the existence of two regions, low rate at low temperature and high rate at high temperature, with a discontinuity between the two, but with the same activation energies in both regions. The discontinuities in the rate curves are particularly sharp when finely ground material is used.  $\text{KBrO}_3$  that has been subjected to gamma-ray irradiation ( $\approx 3 \times 10^6$  r of  $\text{Co}^{60}$ ) is characterized by a single decomposition rate region, i.e., the irradiation has eliminated the dis-

continuity between the two regions. Also, once normal  $\text{KBrO}_3$  has been partly decomposed at a higher temperature, it decomposes at low temperature at a rate given by extrapolating the high temperature curve. Explanations for this behavior can be given in terms of classical phenomena such as melting, crystal break-up, and diffusion. Microscopic observations support such an interpretation, since they showed that surface premelting plays an important role in the decomposition.

A series of experiments have been started on the influence of radiation on the simplest solid-gas reaction, namely adsorption of a gas. In one study, the physical adsorption of krypton on unirradiated and neutron-irradiated alumina has been measured. Adsorption was found to increase upon irradiation. Analysis of the data showed that the radiation created high energy sites for adsorption. The behavior of alumina with respect to hydrogen adsorption is quite different. There is no change in the surface area upon irradiation (as measured by nitrogen adsorption), but there is a significant decrease in the hydrogen adsorption. The adsorption isotherms can be fitted by Langmuir's equation and are therefore characterized by two parameters: the probability of adhesion to the surface, and the density of molecular packing in the adsorbed monolayer. The data available at present indicate that, upon irradiation, the adhesion of the gas molecules is increased but the density of packing is decreased.

### Structure of Solids

Another major research activity in solid state physics is the study of the structure of solids by neutron and x-ray diffraction. For the past several years the emphasis has been on magnetic and ferroelectric structures. While most of the effort has been concentrated on structure analyses, some studies have been made in which structure has not been the principal consideration. These have dealt with some fundamental questions in the theory of magnetism and the nature of crystal transitions.

Investigations using polarized neutrons continue to yield important information about electron distributions in various materials. Recent studies have been concerned with iron. The magnetic form factor,  $f$ , of ordered  $\text{Fe}_3\text{Al}$  has been measured out to  $\sin \theta / \lambda \approx 0.7$ . The observed values for  $f$  in this alloy show systematic deviations from the smooth single-valued function appropriate to a spherically symmetric density of the unpaired  $3d$



electrons. The deviations show also that the density is different around the two types of iron atoms found in this superlattice, one having only iron near neighbors and one having half iron and half aluminum near neighbors. Measurements are also in progress on the form factor of a magnetite ( $\text{Fe}_3\text{O}_4$ ) single crystal. Results obtained so far indicate that (1) the charge density for the iron ion is more radially extended than that for iron in the metallic state; (2) there is a slight but definite difference in the form factors for the iron ions on the two types of spinel lattice sites, the radial charge density being slightly more compact on the octahedral than on the tetrahedral ions.

The magnetic structure of  $\text{Mn}_4\text{N}$  has been determined from neutron diffraction data. The cubic unit cell has manganese at the corner and face center positions, and nitrogen at the body center of the cell. Standard powder diffraction techniques led to four possible spin arrangements, but it was possible to eliminate all but one of these by polarized beam studies. Moments of  $\approx 0.8 \mu_B$  and  $3.5 \mu_B$  were found for the face center and corner manganese, respectively. The face center moments are all parallel to each other and antiparallel to the corner moment. As also observed in earlier work on  $\text{Fe}_4\text{N}$ , the results disagree with recent theoretical discussions on compounds of this type, in which an antiparallel arrangement of spins is predicted for opposite face center positions of the cell, although the  $\text{Mn}_4\text{N}$  results differ from those for  $\text{Fe}_4\text{N}$  in some other respects. Instead of the parallel alignment observed in  $\text{Fe}_4\text{N}$ , the face center moments are opposed to the corner moment. Also, the observed moments in  $\text{Fe}_4\text{N}$  are explicable by assuming reasonable electronic configurations and by applying Hund's rule, but the small face center moment of  $0.8 \mu_B$  in  $\text{Mn}_4\text{N}$  cannot be obtained in this way. Hund's rule can be satisfied for the corner moment, however, if a configuration of  $3d^{6.4}4s^{0.6}$  is assumed for this site. One then obtains  $5.0\uparrow d$  and  $1.5\downarrow d$  electrons. The  $\text{Fe}_4\text{N}$  results, and also some magnetic measurements on similar compounds, indicate rather strongly that nitrogen effectively donates an electron to each of the face center  $d$  shells in bonding. If on this basis a configuration of  $3d^{7.4}4s^{0.6}$  is assumed for the face center manganese, the low moment can be accounted for by the distribution  $4.1\downarrow d$  and  $3.3\uparrow d$  electrons.

Antiferromagnetism has been observed in several orthorhombic  $\text{CrVO}_4$ -type structures by low temperature neutron diffraction measurements.

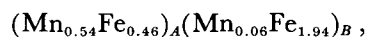
The crystals examined so far are  $\text{CrVO}_4$ ,  $\text{NiSO}_4$ ,  $\text{FeSO}_4$ , and  $\text{MnSO}_4$ . The anomalous magnetic properties of the anhydrous sulfates were discovered many years ago in the course of low temperature studies at the Kammerlingh Onnes Laboratory in Leiden. It is of some historical interest that these compounds were among the first substances found to exhibit what is now recognized as characteristic antiferromagnetic behavior. The magnetic ions are located on simple symmetry-fixed positions in the unit cell:  $0, 0, 0$ ;  $\frac{1}{2}, \frac{1}{2}, 0$ ;  $\frac{1}{2}, 0, 0$ ;  $\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$ . Two basic types of antiferromagnetic structures have been found so far: alternating ferromagnetic sheets on the (002) planes, and alternating ferromagnetic sheets on the (020) planes.  $\text{CrVO}_4$  and  $\text{MnSO}_4$  show the first type of spin ordering, and  $\text{NiSO}_4$  and  $\text{FeSO}_4$  show the second. In both cases, the magnetic unit cell is identical with the chemical cell. An attempt is being made to locate the spin axes, which in principle can be determined uniquely in cases of orthorhombic symmetry; however, this has been difficult to do accurately with powder data. Some of the magnetic interactions are quite unusual, since they must occur in linkages such as  $\text{Fe}-\text{O}-\text{S}-\text{O}-\text{Fe}$ .

Low temperature powder studies have been made to determine the spin structures of the isomorphous carbonates of manganese and iron, which have the calcite structure (rhombohedral). In both cases the spin alignment is antiferromagnetic and corresponds to having the close-packed anion layers alternate in sign along the [111] direction. In the case of  $\text{FeCO}_3$  the spin direction is [111], while for  $\text{MnCO}_3$  it is perpendicular to [111], although there are small deviations in the observed data from this model which may indicate that some departure from completely antiparallel alignment is responsible for the small parasitic moment observed in this compound.

Two other rhombohedral oxides with the corundum structure,  $\text{V}_2\text{O}_3$  and  $\text{Ti}_2\text{O}_3$ , were investigated to confirm the antiferromagnetic orderings suggested by magnetic susceptibility data. In neither case was coherent magnetic scattering observed. It must be concluded, therefore, either that the localized moment in the case of antiferromagnetic ordering is anomalously low, or that the transitions observed in the susceptibility, resistivity, and coefficient of expansion have some other origin.

Work in the spinel system has continued with an investigation now in progress of the cation

distribution and spin structure of  $\text{Mn}_{0.6}\text{Fe}_{2.4}\text{O}_4$ . The cation distribution found is



where  $A$  refers to the tetrahedral and  $B$  to the octahedral sites. Moreover, a weak superstructure line suggests a 1:1 ordering of manganese and iron on the  $A$  sites, which may be useful in explaining the unusually small magnetic anisotropy in this compound.

Studies on the ferroelectric crystal triglycine sulfate have continued. Neutron data collection for a  $b$ -axis projection are now completed, and least-squares calculations are under way (present reliability index = 16%). This will be the second projection of this structure and should complete the analysis. As part of the investigation of the nature of ferroelectricity in this compound, x-ray diffuse scattering studies have been made as a function of temperature, and critical scattering was observed at the Curie point. The experimental data are consistent with a statistical treatment of the transition as an order-disorder problem.

Considerable progress has been made on a study of the structural nature of ferroelectricity in the closely related salts  $(\text{NH}_4)_2\text{SO}_4$  and  $(\text{NH}_4)_2\text{BeF}_4$ . The crystal structures of these two compounds are very nearly the same, but ferroelectricity develops along different axes. In the case of the sulfate, an accurate three-dimensional x-ray analysis has been completed for the low temperature phase. One neutron projection is still needed in order to obtain an accurate set of hydrogen coordinates.  $(\text{NH}_4)_2\text{BeF}_4$  has been examined at room temperature in a three-dimensional x-ray analysis and by projections with single-crystal neutron data. This work is almost complete, but some disorder in the structure is still not understood.

The validity of certain theoretical explanations of the ferroelectric behavior of  $\text{KH}_2\text{PO}_4$ -type ferroelectrics has been under study. Specifically, a model has been proposed in the literature recently that predicted an asymmetric double potential well for the  $\text{O}-\text{H} \dots \text{O}$  bonds below the Curie point. A careful least-squares analysis of available data indicates, however, that the proton is in a single-minimum asymmetric well. The nature of the hydrogen ordering transition is being investigated by looking for critical neutron scattering in  $\text{KD}_2\text{AsO}_4$ . Critical scattering has been observed

by examining the background near the (600) Bragg reflection as a function of temperature. This first experiment was a crude one with poor statistics, however, and the small peak obtained must be checked with more refined techniques.

The structure of orthorhombic  $\text{PbO}$  was determined from neutron powder diffraction data. The lead positions had been determined fairly well in an earlier x-ray study, but the oxygens could be placed only on the basis of what appeared to be a "logical" structure. The space group reported as most likely was  $Pb2_1a$ . The neutron analysis showed that the correct space group is  $Pbma$ . Accurate positions were determined for both lead and oxygen.

Machine analysis of the short-range-order diffuse scattering of monochromatic neutrons measured in both the [100] and [111] directions of  $\beta$ -brass single crystals at  $550^\circ\text{C}$  is nearing completion. The short-range-order parameters obtained from the least-squares analysis are a quantitative measure of both the degree and extent of the local order. From the combined [100] and [111] data it is theoretically possible to solve for the first 23 short-range-order parameters. In most systems measured to date, the local order has been found vanishingly small for atoms more than 5 to 10 neighbors apart. This is not the case for  $\beta$ -brass, and the most important result of the machine calculations is that order persists over distances greater than those for which it is possible to solve from the [100] and [111] scattering data. In view of the low thermal flux of the Brookhaven Graphite Research Reactor, the time needed to collect the additional data required for a more complete analysis would have caused serious changes in the composition of the crystals maintained at  $550^\circ\text{C}$  by the evaporation of zinc.

A sufficient quantity of the isotope  $\text{Cu}^{65}$  has been received from the Isotope Division of Oak Ridge to grow a single crystal of  $\alpha$ -brass large enough for neutron diffuse scattering studies. Only a study of the scattering from such an enriched single crystal, annealed to produce a favorable degree of short-range order, can give conclusive evidence of local order. Several unenriched crystals have been grown, essentially to develop a technique to reduce porosity and lineage usually found in single crystals of  $\alpha$ -brass. Porosity is undesirable, since it can produce scattering similar to that characteristic of clustering.

# Accelerator Development

## ALTERNATING GRADIENT SYNCHROTRON

The major effort of the Accelerator Development Department continues to be the construction of the Alternating Gradient Synchrotron (AGS), on which work was begun in 1954. Today, some six years later, construction of all major components has been completed, and the preliminary trials of the machine are in process.

Figure 1 shows a site plan of the AGS. This complex consists of four major completed structures and a fifth in the planning stage. The Service Building houses the offices, laboratories, machine shop, electronic shop, and the power and water centers. The Linac Building contains the Cockcroft-Walton preaccelerator and the linear accelerator injector for the synchrotron. A reinforced concrete tunnel 18×18 ft in cross section and  $\approx\frac{1}{2}$  mile in circumference encloses the machine proper. The Target Building and outdoor experimental area will contain the facilities, beam paths, and auxiliary equipment required for the utilization of the AGS. An additional enclosed experimental area (east area) is being designed and will be available to meet the growing requirements of the research program.

The AGS preaccelerator is a 750-kev Cockcroft-Walton generator. Protons for the machine are derived from a cold cathode ion source located at the

top of the filter stack of this power supply. The 750-kev beam is injected into a 110-ft-long linear accelerator and is accelerated to 50 Mev. The linear accelerator (linac) has 124 contoured drift tubes appropriately spaced along its length, and the system is resonated at  $\approx 200$  Mc. Focusing is accomplished by means of quadrupole magnets enclosed in the drift tubes. An array of magnets has been placed between the high energy end of the linac and the synchrotron ring to bend, translate, focus, and analyze the beam as required. At the conjunction section of the injector and synchrotron, a pulsed electric field removes a  $1.5^\circ$  angle to launch the beam on a satisfactory orbit.

The ring is composed of 240 magnets, 60 correcting magnets, 12 accelerating stations, and various straight sections arranged in a circle  $\approx\frac{1}{2}$  mile in circumference.

The 240 magnets have been designed to provide a uniform azimuthal field to bend the beam, and a radial gradient (which alternates every two magnet units) to provide focusing in both the horizontal and vertical planes. Quadrupole and sextupole magnets have been designed and positioned around the ring to correct the saturation behavior of the main magnets. Power to the main magnets is supplied by a 12-phase alternator rated at 36,000 kva driven by a 6000-hp wound rotor induction motor. A 47-ton flywheel mounted on the common shaft provides additional energy storage mass to the system. The alternator feeds a bank of 24 ignitrons that rectify the power to deliver 6000 v and a current pulse that rises from 0 to 6500 amp in  $\approx 1.2$  sec. The ignitrons also serve to recover the energy stored in the magnetic field by inverting and driving the alternator as a motor to store the energy again in the total rotating mass.

Acceleration of the beam from 50 Mev to 30 Bev is accomplished by means of 12 rf power amplifier accelerating stations. In passing each of these 12 locations the particles gain 8 kev, or  $\approx 100$  kev per revolution. The proper drive for these power amplifiers is derived from the beam itself. These low level signals are amplified and phase-corrected in a complex electronic system. Each signal then is further amplified in a high-powered

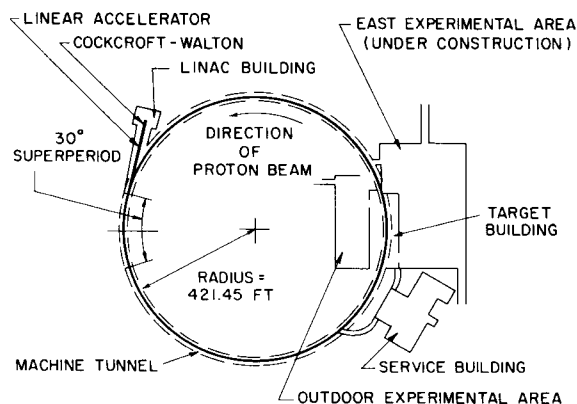


Figure 1. Site plan of the Alternating Gradient Synchrotron complex.

central driver that feeds all accelerating stations in the magnet enclosure.

The vacuum chamber of the synchrotron is fabricated of Inconel-X (a high resistivity material), 0.078-in. thick, of elliptical cross section with a 3-in. vertical axis and a 6-in. horizontal axis. Evapor-Ion pumps are used throughout the system, including the linear accelerator. These devices pump through the gettering action of evaporated titanium and gas ionization. They have proved to be extremely fast and reliable.

The various components of the AGS have been independently tested. To date, only the linear accelerator injector has been extensively operated as a machine. Detailed plans have been made for the first system testing of the entire synchrotron. Many measurements must be made of the machine parameters to explore and understand fully its behavior.

During fiscal 1960, all 240 AGS magnets were set in their precisely surveyed final positions on the ring. The fabrication of the correcting magnets was completed, and after extensive measurements these units also were placed in position. The installation of the main magnet power supply was completed. Load testing of the system presented a number of problems, whose elimination in some cases will require extensive modifications.

The performance of the Cockcroft-Walton injector for the linear accelerator has been continuously improved, and it has been operating when needed during the year. Assembly of the linac was completed early in 1960. A proton beam was successfully accelerated to 50 Mev on April 13, 1960. Subsequent assembly of the lens array between the linac and the synchrotron permitted the beam to be transported to the inflector and launched into the synchrotron.

Construction and installation were completed of all major sections of the radio-frequency system. All components were bench-tested under simulated beam conditions, and after the usual amount of "debugging" no fundamental troubles are anticipated. A fault in the central dc power supply required some rebuilding of the unit. Some circuit additions resulted in an improvement in the output characteristics of the supply. Most of the beam observation electrodes and amplifiers were installed in position around the ring.

The linac vacuum system has been satisfactorily maintaining a vacuum of  $\approx 4 \times 10^{-6}$  mm Hg. Installation has been completed of the synchrotron

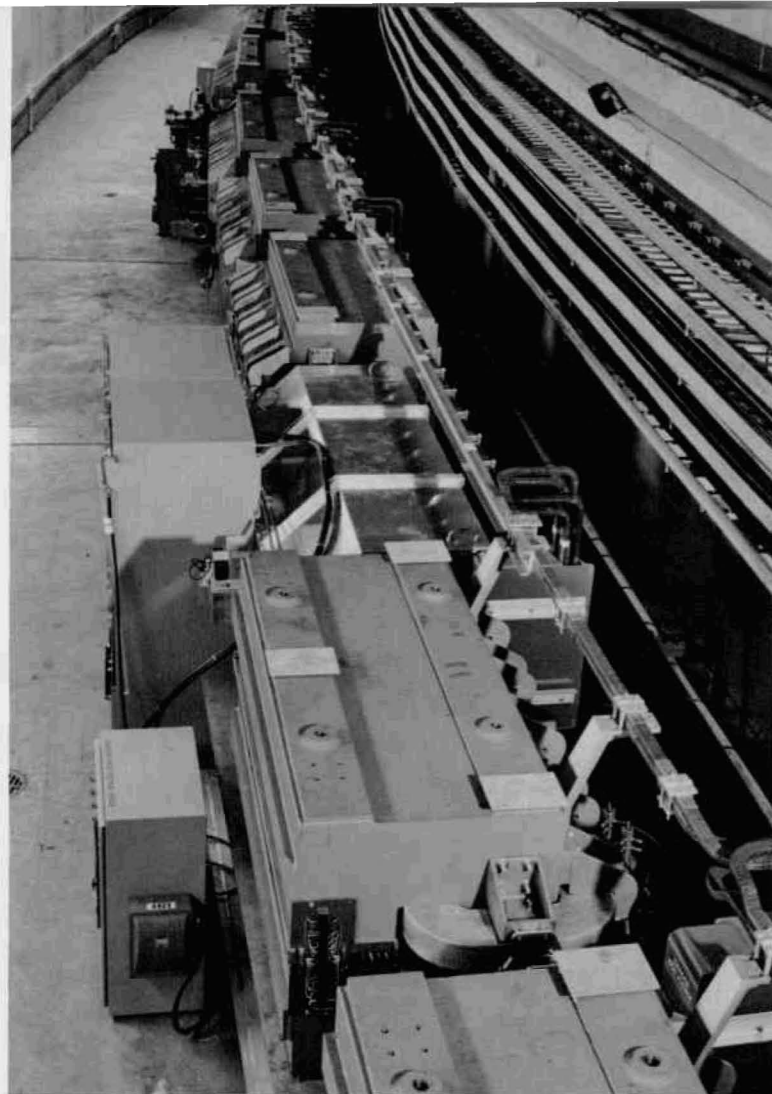


Figure 2. AGS tunnel showing typical arrangement of magnets, accelerating station, pumping station, survey monument, and cable trays.

vacuum chamber, Evapor-Ion pumps, and auxiliary equipment. The system was placed in operation superperiod by superperiod. When most of the leaks were eliminated, an average pressure in the  $10^{-6}$  mm Hg range was obtained around the ring.

Most of the AGS controls necessary to operate the linear accelerator and the major synchrotron components have been installed. These controls cover the basic functional and safety aspects of the machine operation. Refinements to the controls have been designed and will be installed as soon as practicable.

On May 17, 1960, a 50-Mev proton beam was injected into the synchrotron and successfully completed one turn around the ring. On May 26, 1960, a 50-Mev proton beam was injected into the synchrotron and successfully spiraled around the machine  $\approx 100$  times.

The number of personnel remained substantially the same during the year. Members of the staff presented papers at the 1960 Institute of Radio Engineers Convention, the American Institute of Electrical Engineers' Fall General Meeting, and the International Conference on High Energy Accelerators and Instrumentation, CERN, 1959.

A total of 25 physicists and engineers from 8 foreign countries visited the Department to discuss mutual problems.

Details of design and construction of the AGS components are presented below.

### **Magnet**

The crucial test of the entire magnet system occurred during the injection trials of the whole synchrotron. At the first attempt, the injected beam successfully made a complete turn, and, at the first attempt with the pulsed inflector, the beam spiraled for the correct number of turns, which indicated that there were no serious perturbations in the magnetic field. These successes climax a long program of computing, testing, measuring, and surveying.

Since all the main magnets were in their approximate positions on the ring at the end of the last fiscal year, further construction work has involved only clean-up operations and the addition of protective devices such as coil covers and end boxes. The data from the magnetic measurement program were used for surveying the magnets and placing them in their final locations.

The preliminary adjustment of the magnets to their specified positions on the ring was completed in September 1959. A precise traverse of the 24 control monuments and a level circuit of the bench marks on the pile caps supporting the magnet girders were begun in October, and another series was initiated in January 1960. These surveys indicated that the foundations were stable within 0.010 in. per year unless the loads were changed. The soil reacts elastically with changes in the loads. Thus the girder supports at the north end of the Target Building settled 0.091 in. with the placement of 1400 tons of shielding to seal off the openings there, while the south end rose 0.020 in. with the removal of concrete top beams, which had been stored there until needed at the north end. The subsequent placing of 560 tons of shielding to seal the south end caused a maximum settlement of 0.019 in. in the adjacent girder which gradually tapered off and disappeared at a distance of  $\approx 80$  ft.

The data from the January-February precise surveys were used to realign the magnets radially and vertically immediately prior to the final assembly of the vacuum chamber. The changes required to realign the magnets (except those that had been moved for overhaul of supporting mechanisms) were generally within  $\pm 0.010$  in. A recent leveling showed no appreciable changes with the exception of a rise of 0.017 in. in a section  $\approx 100$  ft from an excavation for the site of the proposed Bubble Chamber Building. Because of such sensitivity to changes in loading, check surveys will be made before and after construction and experimental equipment installations.

The 24 quadrupoles and 36 sextupoles were delivered, measured mechanically and magnetically, and installed in their appropriate locations on the ring. Small differences (of the order of a few mils) between the mechanical and magnetic centers were taken into account during the precise positioning of the multipoles. In the quadrupoles, there was no indication of a dipole term that might have been introduced by the splitting of the magnet yoke, but in the sextupoles the measurements showed the existence of a dipole term of  $\approx 0.05\%$  of the sextupole term at the maximum 3-in. radius. Remanent fields in the quadrupoles were of the order of 5 gauss, and in the sextupoles were  $\approx 3$  to 4 gauss. Since heavy cable to connect the multipoles to their generators had not been installed in time for the synchrotron's injection trials, lightweight wiring was connected in a temporary installation to supply  $\approx 5$  amp maximum current, which was sufficient to provide the small corrections necessary at injection fields.

Small windings were also placed on the back-legs of all the main magnets and connected in such a way as to provide both sine and cosine terms for the 7th, 8th, 9th, and 10th harmonics in the injection fields in case it became necessary to correct for harmonic perturbations in the main magnetic field. For the same reason, 6-in.-long kicker coils were added in 96 straight sections to provide the sine and cosine terms of the 8th and 9th harmonics in a radial field. These coils could move the median plane  $\approx \pm 0.200$  in. at injection. However, the coils were not used during the injection and acceleration trials of the synchrotron; they will probably be used only for studies of beam dynamics.

The main magnet power supply was pulsed for the first time on August 20, 1959. Since that time,



considerable difficulty has been experienced with the equipment. The problems ranged from a minor lubricating problem to those requiring major component redesign, including an axial motion of the rotor assembly under pulsed operation and inadequate performance of the liquid rheostat motor control system. The axial motion has been successfully contained by a thrust bearing mounted from a new, fabricated steel, bearing pedestal. As yet, the motor control system has not performed adequately, and a completely redesigned servo system and auxiliaries are ready for installation and test.

Delivery has been made and installation completed of the motor generator power supplies for the correcting lenses. Design of controls for these units is under way. However, these power supplies will not be required for the initial beam acceleration test.

### Linear Accelerator

The linear accelerator and injection system have been completely assembled and tested. Satisfactory proton beams were injected into the synchrotron and were observed to circulate as expected. Although many improvements and modifications are contemplated, performance of the injection system is adequate for initial tests of acceleration in the synchrotron.

A year ago, the linac tank sections were in place but virtually no drift tubes had been completed and located. Construction and hanging of the drift tubes were completed in October 1959, and tuning of the individual tank sections was begun. Because of the close spacing of normal modes of oscillation of the long tank, it was thought that tuning of the tank as a whole would be extremely difficult. For this reason, each tank section is terminated midway between drift tubes and can be tuned by closing its ends with metal end-plates. The computed drift tube shapes gave approximate tuning without adjustment. Final tuning was done by radial motion of a number of "ball tuners" whose positions are controllable from outside the tank. To bring all the tuners close to the tank wall and safely away from high field regions, each tank section was tuned roughly by insertion of a machined copper bar along the tank wall. The tuning of individual sections gave the correct dimensions for these tuning bars. By the end of 1959 these procedures had been completed, and the linac was

assembled in its final configuration, ready for the fine adjustment of the ball tuners to give the "flat" field pattern necessary for acceleration. By this time it was evident that the pattern achieved was close enough to flatness so that final tuning could proceed.

During assembly of the linac, step-by-step testing of the rf power system was undertaken. The first French Thomson-Houston (FTH) amplifier was excited by the 20-kw driver and supplied power to a dummy load. When it had been satisfactorily tuned, it was used to power the second FTH stage, which was then connected to the two parallel FTH amplifiers that drive the linac. The output of the two final stages is combined in a waveguide hybrid junction. For first tests the hybrid junction was terminated by an electromagnetic horn whose impedance simulated that of the linac. Since this system includes some 15 tuning adjustments, a rather tedious step-by-step procedure was evolved which finally resulted in relatively satisfactory impedance matching at all points.

Evacuation of the linac proceeded more slowly than expected. With 20 Evapor-Ion pumps, the pressure was brought rapidly to the  $10^{-5}$  mm Hg range, but great difficulty was experienced in reaching the next order of magnitude. It was considered initially that a pressure of  $2 \times 10^{-6}$  mm Hg would be a desirable operating range, but the combined effects of degassing and of numerous troublesome leaks held the pressure at a figure 4 to 5 times higher than this.

Last March it was decided to attempt powering the linac in spite of the unsatisfactory vacuum. Surprisingly enough, the linac accepted power with only a minor amount of voltage breakdown. The power level was raised over a period of three weeks, and on April 13th the first attempt was made to accelerate a proton beam. This attempt was entirely successful. Without further adjustment a beam was accelerated and was shown to have the design energy of 50 Mev. The machine was found to be relatively insensitive to preinjector energy and to settings of the focusing quadrupoles in the drift tubes. First tests yielded accelerated beams of the order of 1 ma peak.

After the first operation, performance of the linac deteriorated because of rf breakdown at the low energy end, which was traced to leaks in low temperature solder joints in the drift tubes. The tank was opened, and examination of the drift



tubes revealed considerable erosion of these joints in the first ten drift tubes. After some deliberation, the first 15 drift tubes were removed, and the joints were rebuilt and then covered with a dense copper plate. This process seems to have eliminated the erosion problem, but it resulted in drastic detuning of the low energy end of the linac, and extensive retuning procedures were required before 50-Mev operation was again possible.

Operation at 50 Mev is now on a regular basis. Currents of up to 3 ma have been delivered to the injection system in beams whose energy spread is  $< 1$  Mev.

Performance of the preinjector is now routine. Initially currents of 20 to 30 ma were delivered by the cold cathode ion source to the 750-kev pre-accelerating system. In operation, however, these beams had an erratic, high-frequency time struc-

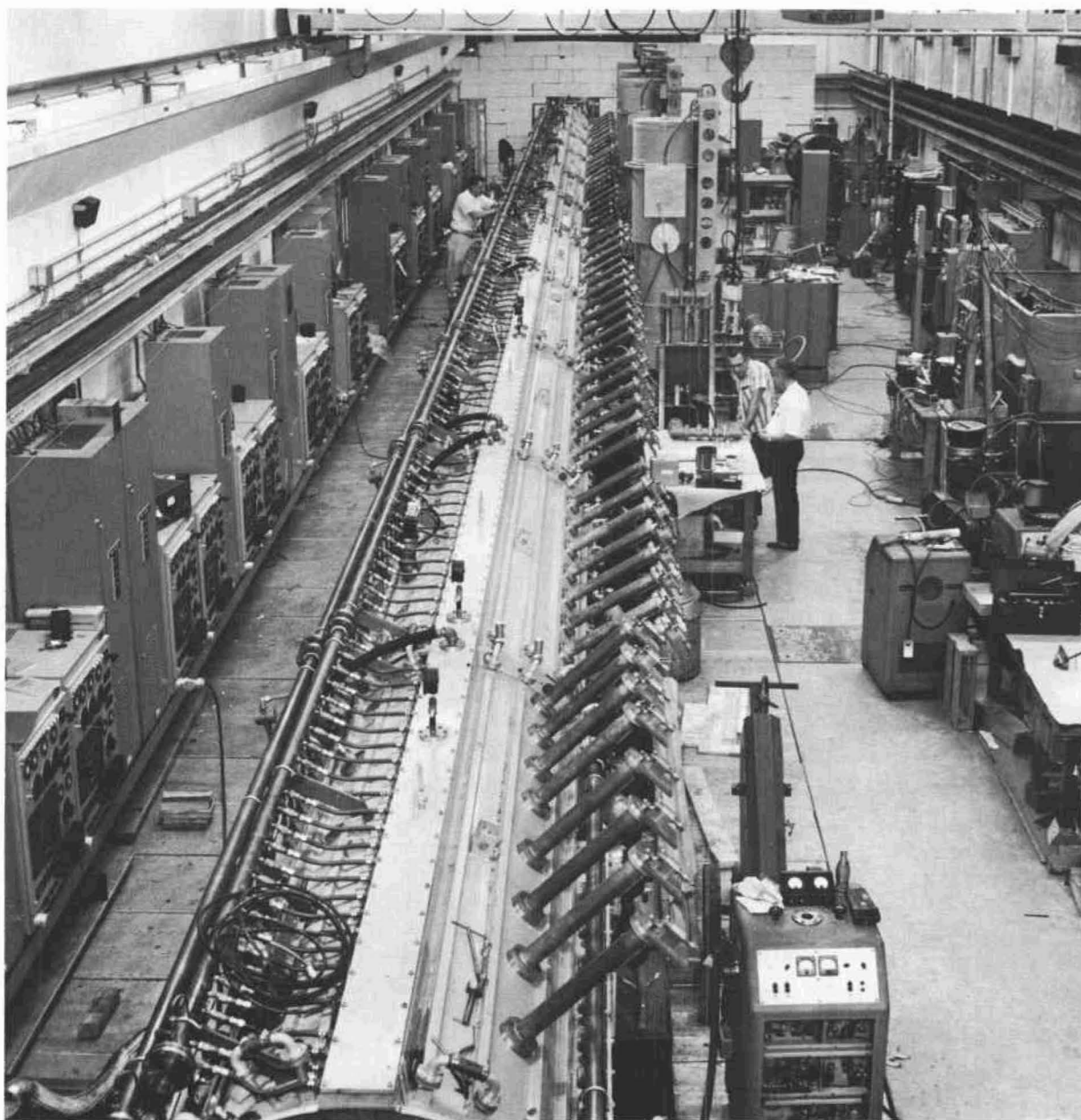


Figure 3. View of the linear accelerator looking from the Cockcroft-Walton machine toward the high energy end.

ture which, so far, it has been possible to control only by reducing the beam intensity. Experiments on other ion source types are in progress, whose purpose is to achieve higher beam intensity. A further increase in intensity of the 50-Mev beam should result from inclusion of a "buncher" between preaccelerator and linac. The buncher has been constructed but not yet installed.

The 140-ft run from the linac to the injection point includes 7 focusing quadrupoles, 8 steering magnets, 1 beam-analyzing magnet, and 3 viewing boxes with beam-defining apertures and movable screens for remote TV examination of the beam. All these items together with vacuum piping, auxiliary equipment, and remote controls were

ready by May of this year. The inflector itself, whose two deflecting plates must be charged to plus and minus 80 kv and then discharged in  $<1$   $\mu$ sec when the beam has made one complete revolution of the synchrotron, was also ready and tested. Consequently, it was possible in May to inject first a single turn and, a week later, a beam that circulated  $\approx 100$  times around the synchrotron. In the absence of rf acceleration in the synchrotron, this approximates the maximum circulation possible before the rising synchrotron field forces the beam to the inner wall of the vacuum chamber. The parameters for focusing and direction of the beam through the complex injection system have been computed, and the procedures

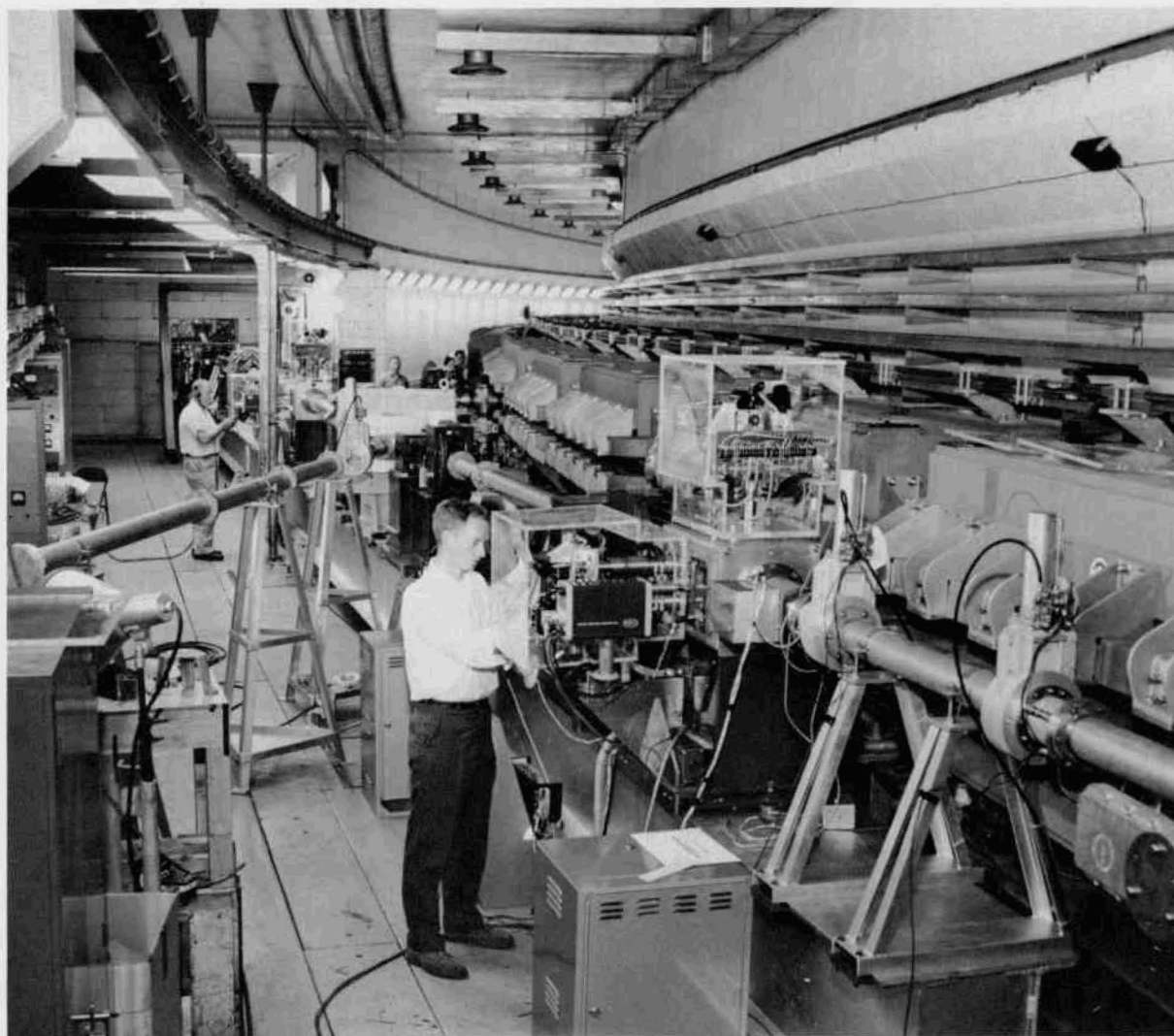


Figure 4. General view of the AGS injection system.

for converting the linac beam into a configuration acceptable by the synchrotron have been developed in great detail. In operation the computed orbits have been extensively verified.

A concrete shielding wall has been installed at the high energy end of the linac. Radiation at this point is rather high, particularly when the beam is intercepted by one of the remote-viewing means. The low energy shield wall has not been erected, since the radiation level here is satisfactorily low. High x-ray emission, often found in proton linacs and associated with minor contamination of drift tube surfaces, has not been observed in this machine, probably because of the efficient Evapor-Ion pumping system and the extensive cleaning procedures used in preparing the linac for evacuation.

### Radio-Frequency

All the major components of the radio-frequency system have been completed and installed. The 12 accelerating stations together with the ferrite cavities, cavity shields, power amplifiers, saturating supplies, and water heat exchanger units were placed in their final positions. These stations have been tested while being fed with an rf signal from the main driver. Problems that were anticipated but failed to materialize include strong interactions among the 12 stations, rf leakage from the cavity structure, and parasitic oscillations in the driver or power amplifiers.

The ripple content of the Moloney Electric Company power supply that feeds the power amplifier and the main driver was found to interfere with proper operation. In addition, the low output impedance of this supply could result in large surge voltages appearing at the ends of the dc feed cables to the amplifier stations. To reduce these effects, an inductance-capacitance filter using one of the early models of the AGS magnet and its coil as the choke was added to the power supply, and no further difficulties have been encountered.

The special 10-ft pickup electrode straight section to supply the radial and sum signals for the low level rf system was constructed, tested and aligned, and subsequently installed in the ring. The necessary signal cables have been run to the control room, including the special low attenuation cable carrying the wide-band sum electrode information. The radial pickup system was modified to allow the electronic equipment to be installed in the control room rather than in the ring

as had been planned originally. The other racks of the low level system are also in position in the control room. The 36 observation pickup electrode structures are now mounted in position in the ring.

The racks containing the electronics for these observation pickup electrode stations were fabricated and assembled by an outside manufacturer; 26 of them are in position and have been tested. All the signal cables have been balanced, matched, and terminated at a patch panel in the central control room.

The basic components of the AGS timing system are installed and connected in the control room.

### Controls

In general, most of the various control systems of the AGS have been installed and are operational. During the fiscal year more than 2,000,000 conductor feet of control cable and 300,000 feet of coaxial cable have been installed. In addition, various power circuits have been extended as required. Many individual installation arrangements have been devised for machine components, including extensions to the existing general cable tray system, local junction boxes, power and control distribution centers, local power disconnects, and special cable routing.

Both the linac control room and the main control room in the Service Building are in operation. The general arrangement of equipment in these rooms was worked out, including the location of removable floor sections for maximum flexibility in making modifications. Much of the final equipment has been installed, although many of the details are provisional until sufficient operating experience has been gained.

Below are brief comments on some of the control systems installed to date.

A central control panel in the main control room permits master operation and provides information on the over-all system status of the high level rf system, including the central driver, the 12 power amplifiers, and the central dc power supply. Operating experience of the past several weeks has indicated that this control system should be reviewed and that some of the interlocking, sequencing, and protective circuits need revision.

Control of the ring vacuum system and its 75 Evapor-Ion pumps is decentralized except for a few centrally located temporary controls. A three-rack operating and read-out panel is being wired for installation in the main control room.

Circuits for controlling ionization gauges for measuring high vacuum were designed, and prototypes were built and tested. It was then found that manufacture of the magnetic amplifiers around which the circuits were designed was to be discontinued. Consequently the completely static device approach had to be abandoned. Commercial ionization gauge chassis using vacuum tubes were then modified to provide more complete internal interlocking and to permit remote read-out and range change.

In general, the linac controls are complete; the only work remaining is the installation of 150 system status indicator lights on the main console. A modification to the linac vacuum system has been designed and is being installed to provide closer interlocking of Evapor-Ion pumps and their high vacuum valves and controlled sequencing of operation.

A master control panel permitting the simultaneous turn-on, gain change, and mode selection of the 36 pickup electrode stations is in operation.

A temporary, but at present adequate, timing system with a distribution patch panel has been provided in the main control room.

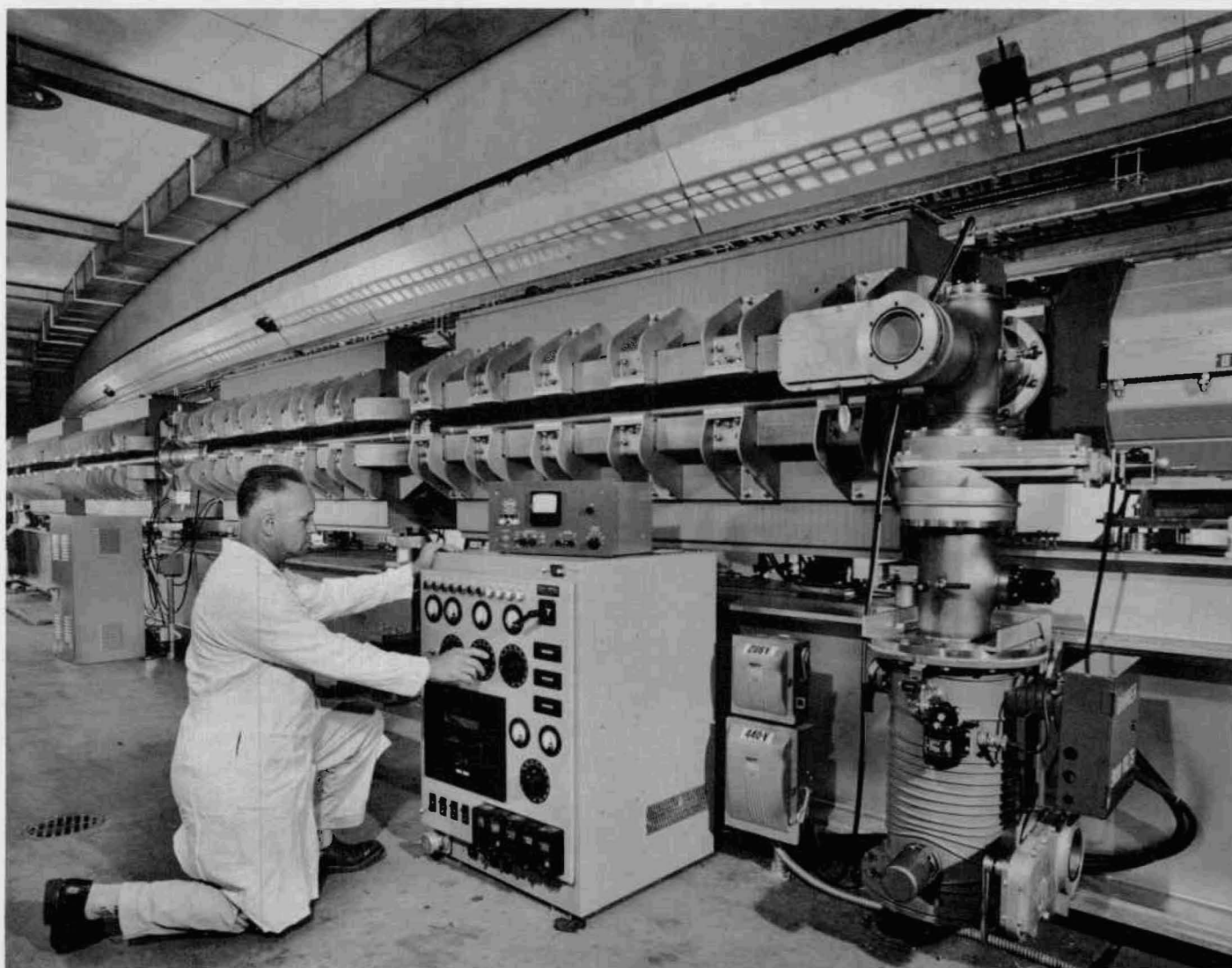
For remote positioning of aperture flags, circuits incorporating balancing relays have been designed, tested, and installed. This system permits the use of calibrated knobs for accurate setting of the flags to intercept the proton beam as required.

A closed circuit television system including 11 cameras and 6 monitors has been procured and modified and is being used for remote viewing of proton beams on screen-wire and quartz flags. Remote operation of the flags themselves has also been achieved.

The installation of the various communications systems (public address, intercom, and interphone) is in its final phase. Many parts of these systems are already being used in connection with operation of the AGS.

Controls and monitoring devices for the water and air-cooling systems associated with various parts of the AGS are now complete and are per-

Figure 5. Evapor-Ion pump and control console in position on the synchrotron ring.





forming according to the original designs and specifications.

### **Vacuum**

The linear accelerator vacuum system was initially placed in operation in February 1960. After an extensive leak detection program to eliminate most of the leaks, and continuous pumping to outgas the surfaces, the pressure in the linac is consistently at  $\approx 4 \times 10^{-6}$  mm Hg. The Evapor-Ion pumps have provided an essentially clean tank which requires very little conditioning time for bringing up the rf voltage. These pumps are now running on a 4-hr ion pumping, 15-min titanium pumping schedule. As a consequence, the maintenance program has been less extensive than anticipated from earlier experience.

The ring vacuum system was placed in operation in May 1960. All superperiods are running on a 12-hr ion pumping, 15-min titanium pumping schedule. The pressures around the ring vary from  $6 \times 10^{-7}$  to  $2 \times 10^{-6}$  mm Hg. The sections at the higher pressures contain materials that have recently been introduced, and the outgassing rate is still high.

Several accidental exposures to atmospheric pressure have occurred in both the linac and the

ring. These exposures resulted merely in inconvenience instead of the catastrophe possible if conventional diffusion pumping systems had been employed.

### **Structures**

Early in 1960, construction was completed on the office addition to the Service Building, and this area was occupied by Department personnel. In March 1960 negotiations were concluded with the Stone and Webster Engineering Corporation, and their contract was extended to cover the design and supervision of the construction of the 58,000 square-foot experimental area on the east side of the Target Building and a 9600 square-foot laboratory addition to the Service Building. Extensive modifications of the electrical and mechanical services in the Target Building have been undertaken to permit the use of hydrogen in this area.

The high iron content of the water produced by the existing AGS wells continues to be a persistent problem. Detailed studies of the aquifers at various levels were made by means of test wells. The data obtained from this program showed that the iron content of water pumped from depths  $>600$  ft was still too high.

# Instrumentation

As the instrumentation required for scientific experiments becomes more complicated, its design and maintenance tend to become a larger fraction of the research effort. This trend is reflected in the growth of the Instrumentation Division. The staff was augmented by 3 engineers and 6 technicians this year. A group has been formed to construct circuits in small quantities and on short notice for high energy accelerator experiments.

For the past three years construction of the digital computer Merlin has been the major project of this Division. This year the computer was brought into operation and turned over to the Applied Mathematics Division; however, maintenance and further improvements will continue to be supplied by this Division.

The major emphasis this year has been on ultra-high-speed circuits for use at the accelerators. These circuits include pulse amplifiers, electronic counters, and circuits for time measurement at very high pulse rates and with resolution comparable to the time it takes light to travel one foot.

Another activity of increasing importance is data handling, which involves automatic systems for data taking, time or amplitude sorting, automatic plotting, and similar procedures. Some experiments can be carried out only with the help of highly automatic data handling circuits. In other cases the instrumentation provides greater reliability or an important saving in manpower.

A third major category of activity is instrumentation for the measurement of small direct currents. One example is a considerably improved circuit for polarographic measurements, a technique used in chemical analysis. Another example is a highly sensitive portable radiation monitor for use in the vicinity of pulsed accelerators.

One member of the Division has spent the year working on solid state radiation detectors in cooperation with Bell Telephone Laboratories, Murray Hill, N.J. This cooperative program has produced improved detectors and pulse amplifiers designed for use with them. The solid state detectors, which are somewhat similar to silicon junction diodes, hold great promise for research and commercial applications.

As the Laboratory grows larger, the requirement for repair and calibration of instruments also increases. The number of technicians assigned to this work increased from 6 to 9 during the fiscal year. The calibration facility that has serviced all the Health Physics instruments for the past 12 years was torn down to make room for the new Physics Building. A new location was provided for this activity near the Meteorology Building.

In addition to the major new developments in the field of instrumentation, described briefly below, a variety of components and systems of established design were built.

## MILLIMICROSECOND CIRCUITS

Counter experiments at the large accelerators require very high-speed circuits. Last year a start was made on the design of transistor circuits for this application. A multicoincidence circuit suitable for operation with pulses of one or a few nanoseconds ( $10^{-9}$  sec) in duration was described in the previous annual report. This circuit has been improved, and a number of other "building blocks" have been designed, including a pulse amplifier, a fast scaling circuit, and a "fan-out." The amplifier consists of 3 transistors and 2 pulse transformers. Primary and secondary windings of the transformer are wound as a twisted pair on a very small ferrite core. The amplifiers have a voltage gain of 10, input and output impedance of 50 ohms, maximum output voltage of  $\frac{1}{4}$  v, and a rise time of 2 nanosec. The binary scaler serves also as a pulse amplitude discriminator and can resolve two pulses 10 nanosec apart. Normally the signal from a photomultiplier can drive only one output cable without being degraded. The fan-out circuit distributes the signal to four 50-ohm cables, while maintaining the original pulse amplitude and speed.

Each of these building blocks is built in a small chassis, with the necessary connectors and controls, so that a substantial number of them may be mounted in a laboratory rack and interconnected with ease. Work is continuing on the design of new building blocks and the improvement of those now

in use. Tunnel diodes have some advantages for this type of work, and their use is being studied.

A related problem is that of a suitable high voltage supply for use with the fast, high gain photomultipliers. Although the average current drain of these tubes is very small, peak currents up to 1 amp may be drawn during a signal pulse, and the rate of occurrence of signals from an accelerator varies over a wide range. It is expensive and hazardous to supply the dynodes from a high current power supply and voltage divider, whereas low current systems fail at high pulse rates. A power supply that can deliver current as required has been built. It consists of a transistor power oscillator which applies a constant ac potential to the primary of a transformer. There is a multiplicity of secondary windings, each with a silicon rectifier and filter, so that there is, in effect, a stable dc power supply connected between each pair of dynodes. There is some gain modulation at the oscillator frequency, but the gain is independent of the variations in pulse amplitude and rate normally encountered in these experiments.

### SOLID STATE DETECTORS

An important new addition to the family of radiation detectors is the solid state detector. K.G. McKay of Bell Telephone Laboratories showed in 1951 that a  $p-n$  junction responded to alpha-particles. Several workers have exploited this discovery. Last year Dr. Walter Brown of Bell Labs. invited Brookhaven to cooperate in the study of these devices. An arrangement was made for G.L. Miller of the BNL Instrumentation Division to work part-time at Bell Labs., Murray Hill, N.J. This venture has produced some excellent experimental detectors and several other interesting solid state devices. Bell Labs. personnel are responsible for the device development. Brookhaven has supplied electronic equipment and measured the performance of the detectors. To date the most useful properties of solid state detectors are the very high energy resolution with heavily ionizing particles and the linear energy relationship. These properties have been investigated for protons, deuterons, and alpha-particles at the 4-Mev Van de Graaff. The response to beta-rays, gamma-rays, and minimum ionizing particles has also been investigated on a limited scale. These detectors are fast, compared to most other types, and their speed of response has been observed on the BNL sampling oscilloscopes.

In order to achieve high resolution or to detect low energy events, a low noise amplifier is needed. A preamplifier has been designed for this application which has a cascode input stage, a cathode follower stage, and negative feedback through a small capacitor to the input. The internal gain of the feedback loop is enhanced by a "bootstrap" connection from the output to the top of the cascode plate resistor. This arrangement gives a good signal-to-noise ratio, linear gain, and comparatively fast response. Negative feedback through a capacitor has the added advantage that the output signal is proportional to the charge in the signal pulse and quite insensitive to the capacitance of the detector, which varies with the applied collecting potential.

### MERLIN

Construction of the digital computer was virtually complete a year ago. After 12 mo of debugging and logical timing adjustment, the machine had become usable at the close of this fiscal year. While the memory system has been found satisfactory, the barrier grid storage tubes used in it have been a source of considerable difficulty. A new design of storage tube has been used which, although better than the earlier version in certain respects, has proved to have an inferior target performance. Most of the tubes in use have small flaws on the target surface. In order to achieve a satisfactory performance, only 4000 bits are being stored per tube at present. After the completion of an error correction system in the near future, it will be possible to store 8000 bits as originally intended. The magnetic tape system obtained on a subcontract is not yet functioning correctly because of defects in the tape transports supplied. All the orders in the Merlin vocabulary have been checked and appear to function correctly with test routines. It is anticipated that the Merlin computer will be fully operational by January 1961.

### DATA HANDLING

A major modification was made on a time-of-flight analyzer operated by the BNL Physics Department at Chalk River. A set of transistor circuits was added so that time and pulse-height measurements may be made simultaneously. The 1024 storage locations in the memory may now be split to provide from 64 pulse-height channels at 16 selected time-of-flight settings to 8 pulse-height



channels at each of 128 time-of-flight settings. Some restrictions have been placed on the choice of combinations for specific applications in order to keep the new circuits reasonably simple and reliable.

A 100-channel pulse-height-to-digital converter which records on punched paper tape has been built to extend the usefulness of a commercial 100-channel analyzer. The new unit is used to record pulse amplitude from low counting-rate experiments on the paper tape. The converter accepts pulses from two detectors, and an additional hole on the tape is used to identify the source of each recorded event. A tape reader was built and attached to the analyzer so that the slowly accumulated data may be speedily analyzed. One expensive analyzer can sort the data from a number of such tape recording units.

Three digitizing systems have been assembled for punching the coordinates of bubble chamber tracks on IBM cards. Additional information may be inserted from a keyboard. The main problems were to punch the information on the cards in the required order and to obtain reliable operation of the system. A similar project was the construction of an automatic (patchboard-programmed) system to control operation of a crystal neutron spectrometer.

#### DIFFERENTIAL POLAROGRAPH

For several years the members of this Division have been working with the radiochemists on a differential polarograph. The final instrument is more sensitive and more stable than previous instruments. Only a small potential is required to collect an ion in a solution on an electrode. The threshold potential is different for different ions. The electrode current is a function of the potential applied to the cell containing solution, and breaks in the current-voltage curve may serve to identify and to measure the concentration of different ions. Often one of the electrodes is a glass capillary filled with mercury. The mercury, by forming droplets that grow in size and drop at regular intervals, produces a continuously clean surface. It also modulates the current as the surface area changes. In the new apparatus the current is measured with a chopper-stabilized sensitive amplifier, and the amplifier output is sampled at a fixed time in the mercury drop cycle. A second circuit measures the difference in the current between successive samplings and thus gives a reading that is proportional to the derivative of the

current-voltage curve when a linearly varying potential is applied to the cell. An electronic circuit is included to supply the linearly varying potential. The integral and/or differential curve may be displayed on a recorder. A wide variety of sensitivities is provided, and several modes of operation are possible.

#### RADIATION MONITORS

An area monitoring system has been built for the Cosmotron Building. There are ten 5-liter ionization chambers, each with an electrometer circuit and meter. All signals are also displayed on a panel in the control room. The amplifiers integrate the charge due to a single pulse of the Cosmotron on a 10- $\mu$ mf capacitor. The full-scale output of 10 v corresponds to  $\approx 70$  mr, or a dose rate of 500 mrem/hr at a pulse rate of 720 pulses/hr, assuming an rbe of 10 for Cosmotron radiation. The feedback capacitors are shorted just before each pulse. The integrated intensity of each pulse is indicated locally and at the control room console between pulses. The intensity is printed on an inexpensive recorder after each pulse. The signal from each ion chamber is also fed to an analogue-to-digital converter, and the digital values are summed in scaling circuits which then integrate the dose over any desired period of time.

A new monitoring system was built for use over the pool in the Hot Laboratory where cobalt gamma sources are used. The circuits use transistors and silicon diodes except for two long-lived electrometer tubes. At a relatively low level of radiation an audio alarm begins to sound, its pitch rising with the radiation level. When the power mains fail, the circuit automatically switches to a "hot shot" battery. The alarm or warning lights are tripped by most component failures or a weak battery condition.

A portable radiation monitor has been designed for use at the Cosmotron or Alternating Gradient Synchrotron. It has a 5-liter, polyethylene-walled ionization chamber. The amplifier uses an electrometer tube and two transistors. Another transistor is used to generate the bias and chamber polarizing potentials. Two small mercury batteries supply power for 1000 hr of operation. The instrument integrates the ion chamber current on a 10, 100, or 1000- $\mu$ mf capacitor. The scale is calibrated to read 100 mrem/hr on one accelerator pulse on the most sensitive scale (rbe=10). The integrating capacitor is shorted manually as desired.

### OTHER MAJOR PROJECTS

A reactor modulator was built for the Reactor Physics Division of the Nuclear Engineering Department. The mechanical driver is an electromagnet and coil similar to a dynamic loud-speaker assembly. A transistor power amplifier supplies current to the driving coil, which is attached by a wire to a small cadmium cylinder located near the center of the critical assembly. The driven cylinder moves in and out of a fixed cadmium cylinder, which varies the reactivity of the assembly. A linear differential transformer is also driven by the coil and provides a signal proportional to the displacement. By means of a servo loop the dynamic system is forced to follow an applied signal. The motion is  $\pm 1$  cm, and the upper frequency limit is  $\approx 100$  cy.

An analogue tape system was assembled for use in the reactor experiments mentioned above. An available audio tape console was rebuilt. A long loop was made in the tape between the reels so that several read-record heads could be placed along the tape. By varying tape speed, head spacing, and loop length, a wide variety of signal delays are achieved. A voltage-controlled oscillator, ratemeter, and negative feedback amplifier are used to convert analogue signals to frequency modulation for recording, and the same circuits are used to reconvert to an analogue output on playback. The frequency range is dc to 100 cy. The major difficulty was to obtain low flutter in the long tape loop.

A precision regulator circuit was designed for Cosmotron magnet power supplies. The magnetic amplifiers originally supplied have been replaced by a chopper-stabilized dc amplifier.

Two precisely regulated power supplies were made for a beta-ray spectrometer purchased by the Chemistry Department. One supplies current to a Helmholtz coil, and the second forces the field in the spectrometer magnet to follow the field in the reference coil. The latter may also be used to cycle the spectrometer magnet in order to reduce hysteresis effects. Each power supply is doubly regulated by silicon control rectifiers and series transistors. The power supplies stabilize the magnetic field to  $\approx 1$  part in  $10^5$ .

A new generating voltmeter was constructed for the 4-Mev Van de Graaff. It is calibrated to read 0 to 10 Mev in steps of 1 kv, with a meter for interpolation. The readings are reproducible to a fraction of a kilovolt.

A circuit was supplied to modulate the Chemistry Department's 2-Mev electron Van de Graaff. A photomultiplier and electronic switch are mounted in the high voltage terminal. The switch is controlled from circuits at ground potential by a light beam. A  $\frac{1}{4}$ -watt neon lamp was found to be the best modulated light source.

A set of ten moderately low level counters was assembled in a console for the Nuclear Moments Group of the Physics Department. The detectors are small volume, proportional flow counters with sample slides. There is an amplifier and scaler for each detector. Clock circuits are provided so that samples may be measured independently in each channel for predetermined times.

### METEOROLOGY

There have been no major changes in either the personnel or the program of the Meteorology Group during the past fiscal year. The staff at present numbers 13 regular employees and 2 visiting scientists. A third is expected to join the Group in the fall of 1960. These individuals devote most of their attention to the Group's research program, which is oriented principally toward the problems of diffusion and deposition in the lower atmosphere, but also conduct a significant number of service activities which are covered in the Technical Operations and Services section of this report.

#### Research Activities

Foremost in research accomplishments during the past year was the completion of a large number of airborne runs in which the Graphite Research Reactor effluent was traced to distances of 60 miles, and various properties of the plume, such as its height, width, and vertical thickness, were measured. In January 1960, the  $\text{Ar}^{41}$  detection equipment was altered from a simple beta-gamma system to a dual device permitting simultaneous recordings of the mixed beta-gamma and of the gamma radiation only. Thus, relatively simple subtraction gives the beta contribution, and therefore a more precise definition of the actual cloud boundaries, because of the short range of beta-particles in air. Other aspects of the technique, as previously reported, include use of a portable stereophonic tape-recording system to record directly the output from the GM tubes. The Group already has sufficient information to define with fair precision the height to which the reactor cool-

ing air plume rises under various wind conditions, and the information currently being collected should soon permit firm conclusions concerning the diffusion of the cloud at great distances.

Considerable progress has been made in the particulate deposition studies, an important part of the program for some time. This work requires simultaneous measurement of the concentration pattern of a particulate cloud at low levels over a test grid and the deposition occurring on the soil and vegetation beneath it. This poses the very difficult problem of sampling accurately the airborne concentration of particles large enough to be deposited in significant amounts on the ground surface. To accomplish both measurements, a dual particulate emission is used, the one consisting of radioactive  $\text{Cu}^{64}$  spheres in one of 5 size groups whose median diameters range from  $\approx 1$  to  $15 \mu$ , and the other of extremely small uranine particles ( $< 0.5 \mu$ ). The uranine particles are completely collected by filters, whereas the copper particles, though collected inefficiently by the filter assemblies, are readily detectable on the ground surface.

Much of the past year has been devoted to the operation of a pilot diffusion-deposition grid consisting of twelve 2-m stakes equipped with filter samplers and various types of deposition plates and collectors on the ground surface. This grid has accomplished its purpose in assuring that the measuring techniques are suitable, and also in establishing that the  $\text{Cu}^{64}$  can be used without danger or annoyance to Laboratory personnel. In addition, the pilot grid has provided much interesting information concerning particulate deposition itself.

Construction of the full-scale grid (a  $100 \times 100$ -m square) was virtually completed at the end of the fiscal year, and at present the 300 samplers, the vacuum system, and auxiliary equipment are ready for routine operation. The field consists of 100 stakes spaced at 10-m intervals, each equipped with sampling heads at 2, 1, and 0.1 m above the ground.

A solution seems to have been found for one of the more difficult problems in the study, namely, the measurement of true deposition on the soil and vegetation rather than that collected by sticky paper, flat plates, and other such devices. A mobile radiation-counting device has been developed which consists of a shielding pig and detector mounted 20 cm above the ground on a light framework supported by two large bicycle wheels. This

device is rolled to various positions on the grid, and the radiation is recorded on a portable tape recorder. Once calibrated for the  $\text{Cu}^{64}$ , this instrument will provide a measurement of the deposition with minimum disturbance of the surface, and correlation of these results with sticky paper collectors should provide an excellent description of the radioactive deposition on the surface of the field.

### Related Projects

Associated with the copper deposition studies is a continued investigation of the dispersion of ragweed pollen, conducted cooperatively with the New York State Museum and Science Service. Late in the summer of 1959, much information was collected on the dispersion of pollen from a prepared ragweed field to the west of the main meteorology installation. Basically, the technique consisted of obtaining measurements from a large number of fixed samplers radiating outward from the pollen field, and comparing these measurements with background measurements of pollen taken elsewhere on the site. It was shown conclusively that the dispersion of pollen in the area surrounding the field is highly local and that the pollen present in the general atmosphere of the area is probably derived from regions of the size of states.

An important development within this program has been the completion of an isokinetic sampler capable of operating in the natural wind flow. This instrument consists essentially of an intake tube oriented into the wind by means of a fin and so shaped as to minimize the deposition of pollen within the tube itself. The air speed through the intake is matched to the natural wind flow by means of a pair of hot-wire sensing devices, one on the intake and one exposed to the natural wind in a similar housing. An electronic control system varies the vacuum source to match the intake speed to the wind speed. Recording of this fluctuating flow rate, basic to a good quantitative measurement of the pollen concentration, is accomplished on a Fischer-Porter transmitting flowmeter.

The cooperative study of the low level wind profile sponsored by the U.S. Army Signal Corps has continued during the past year, and both a theoretical and experimental study of the low level profile have been substantially completed. Perhaps the most valuable contribution in this work

has been the development of a technique for assessing quantitatively the value of various predictors in this complex process. This technique makes it possible to determine the exact degree of improvement offered by the introduction of a particular predictor, and also whether any improvement can be accomplished over and above the simple estimate of the mean value of the wind speed at a particular height above the ground.

Associated with this study, as well as with other aspects of the Group's activity, has been the development of a much improved wind-sensing assem-

bly. The anemometer is similar in appearance to many others, but differs in that it has an unusually light and durable cup assembly built up from epoxy resin and Fiberglas. The wind vane uses the unambiguous potentiometer recording system developed several years ago by the Group, but internal switching is accomplished by means of a light-beam and photosensitive resistor circuit instead of brush contacts. Thus, the vane assembly, equipped with a Styrofoam fin, has extremely low friction and is as sensitive as the anemometer. Both respond to wind speeds of  $\approx \frac{1}{2}$  mph.

# Applied Mathematics

For the past three years, much of the effort of the Applied Mathematics Division has been concentrated on the Merlin computer, which was constructed in collaboration with the Instrumentation Division. Specifically, the Applied Mathematics Division has been responsible for the logical design of the computer as well as for the preparation of its programming system. While the former task found immediate application in the design and construction of Merlin, the latter effort is still continuing

and will not be completed until the computer becomes fully operational. The construction phase of Merlin was completed early in the year, and the computer became available on a limited basis to members of the Division. During fiscal 1960, the computer was first used exclusively for preliminary testing of operations by the members of the Division who had participated in the logical design. As the reliability of Merlin improved, the computer was also used for the development of the



Figure 1. View of the high-speed digital computer Merlin, designed and constructed at Brookhaven. In the foreground are the high-speed printer and manual control console. The vertical racks to the right rear constitute part of the control section.

programming system. However, the operational status of the machine's fast printer and magnetic tape system did not permit full-scale debugging of the programming system. Nevertheless, the use of the computer by the programming staff has been invaluable in developing the various test routines for the detection of computer malfunctions. Good progress was made in work on a library of standard mathematical subroutines. It is expected that Merlin will become available for general Laboratory use in the fall of 1961.

With the prospect of full-scale operation of Merlin in the near future, work was started on the coding of specific problems. These included the preparation of a multigroup reactor code in collaboration with members of the Reactor Theory Group of the Nuclear Engineering Department, and of a Monte Carlo code for the study of high

energy nuclear reactions in conjunction with members of the Chemistry Department.

In areas not directly related to the use of Merlin, work was carried out on the use of finite difference methods for the solution of hyperbolic and parabolic equations as well as on the bounds for eigenvalues for differential equations. Other research involved mathematical studies of muon capture in helium and a study of the general theory of allowed and forbidden transitions in muon capture. In conjunction with the Physics Department, major progress was made on the solution of many-body problems with the help of digital computers.

In response to the general interest of the BNL scientific staff in the theory of distribution, a group of lectures on the subject was presented during the summer.

# Chemistry

The following sections describe researches lying in or related to the Chemistry Department's continuing fields of interest: chemical effects of radiations; chemical effects of nuclear transformations; applications of isotopes; chemistry of isotopes; nuclear chemistry; molecular structure; and solid state. These studies were carried out by the continuing scientific staff, postdoctoral Research Associates, and visiting scientists. The continuing staff and Research Associates numbered 46 and 16, respectively. During the year 45 visitors participated in the work of the Department.

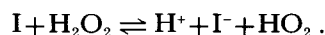
## RADIATION CHEMISTRY

### Aqueous Solutions

The chemical reactions in aqueous solutions under irradiation are produced by short-lived intermediates (free radicals) which result from the excitation and decomposition of water. The observed chemical reactions depend upon the number and the nature of these intermediates, their spatial distribution, and the reactions they undergo with dissolved substances. To obtain information on these points, it is first necessary to establish the sequences of reactions which occur in solutions of different materials. These can then be coordinated to give accurate values of the amounts of different kinds of entities produced. The chemistry of the reactive radicals appears as a ratio of the specific rates with which they react with different compounds, and a table of such ratios constitutes a summary of the chemistry of the particular radical. To place this study on a firmer basis it is obviously desirable to go beyond these relative values by determining absolute reaction rates and lifetimes of these short-lived radicals. The number and kind of radicals obtained in a solution depend in turn on their initial spatial distribution and the resultant competition between diffusion and recombination. Research in this field is therefore proceeding along three lines: (1) determination of reaction mechanisms in a variety of solutions, including the intricate reactions occurring among the decomposition products of pure water itself; (2) special experiments using pulsed radiation or

flow systems designed to determine the lifetimes and hence the absolute reaction rates of the various radicals; and (3) experiments on the scavenging of free radicals at relatively high solute concentrations, in which the details of the nature and initial distribution of the radicals are a predominant factor.

Solutions of iodide ion have been a favorite object of study for many years in radiation chemistry, but many of the detailed phenomena have not been satisfactorily explained. A detailed study of the yields of hydrogen, hydrogen peroxide, and iodine formed under gamma irradiation in dilute iodide solutions has been completed. Irradiation was done in the presence and absence of oxygen, with added peroxide and nitrite, and over a large range of acid concentration. In these solutions, OH radicals oxidize  $I^-$  ions to I atoms, and H reacts with  $O_2$ ,  $H_2O_2$ , or  $I_2$ , whichever is present. The key to detailed understanding of the system appears to lie in the existence of an acid-dependent equilibrium involving atomic I:

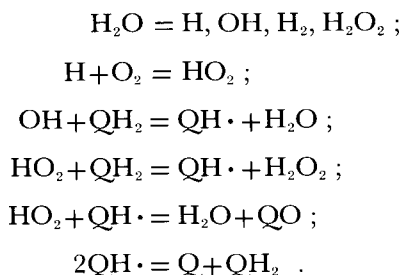


With the aid of this concept, the complicated behavior of the system under different conditions of pH and  $O_2$  concentration may be quantitatively explained. An unexpected result is that in the presence of added iodine the above reaction of I with  $H_2O_2$  does not occur; instead, the full molecular yield of peroxide appears on irradiation of neutral solutions containing added  $I_2$ . The iodine atom, though written above as I for simplicity's sake, is actually believed to be complexed in iodide solution in the form  $I_2^-$ . In the presence of  $I_2$ , it would appear that further complexing must occur, perhaps to  $I_4^-$  or  $I_5^-$ , in which form the iodine is less reactive.

Hydroquinones offer a convenient system for distinguishing between different kinds of radicals. Under irradiation in the presence of air, various substituted hydroquinones are oxidized to quinones with differing yields, depending on whether they are oxidized by OH only, by OH and  $HO_2$ , or by OH,  $HO_2$ , and  $H_2O_2$ . In addition, a small yield of hydroxyquinone also appears. A detailed

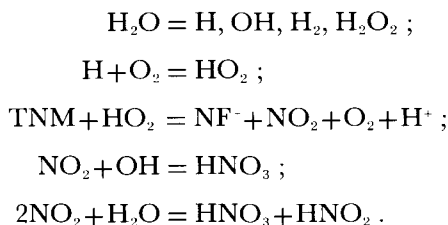


study of the yield of hydroxyquinone has now been carried out in the case of the unsubstituted hydroquinone,  $C_6H_4(OH)_2$ . The products are quinone ( $C_6H_4O_2$ ) and hydroxyquinone ( $HO C_6H_3O_2$ ). These three substances may be called  $QH_2$ ,  $Q$ , and  $QO$ , respectively. The first intermediate in the oxidation of hydroquinone by free radicals is known to be the semiquinone,  $QH\cdot$ . The yields of  $Q$ ,  $QO$ , and  $H_2O_2$ , on irradiation with gamma-rays of  $QH_2$  in air-saturated solution, are found to vary with the concentration of  $QH_2$  and with the radiation intensity approximately as expected from the reaction sequence:



According to this scheme,  $QO$  derives from a reaction between the two free radicals  $QH\cdot$  and  $HO_2$ . Its yield should thus increase with radiation intensity, which increases the concentration of free radicals, and should decrease with increasing concentration of  $QH_2$ , which competes with  $QH\cdot$  for reaction with  $HO_2$ . This expectation has been borne out by the experimental results.

Tetranitromethane,  $C(NO_2)_4$  (called TNM), is an interesting compound whose radiation chemistry has been studied in other laboratories. In dilute aqueous solution it is readily reduced to the stable nitroform anion,  $C(NO_2)_3^-$  (called  $NF^-$ ). The reaction is readily followed, since nitroform has a very high extinction coefficient in the near ultraviolet. More complete and accurate data than those in the literature have now been obtained. The radiation chemistry at  $pH > 2$  can be accurately described by the following simple reaction scheme:



The scheme obviously predicts these product yields:

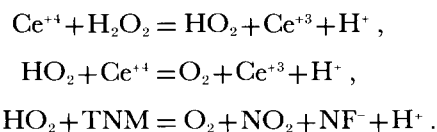
$$G_{NF^-} = G_H ;$$

$$G_{H_2O_2} \text{ (total)} = G_{H_2O_2} \text{ (molecular yield from water)} ;$$

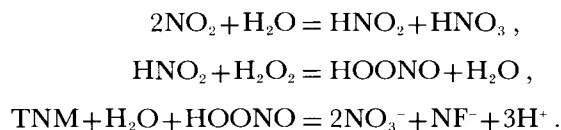
$$G_{HNO_2} = \frac{1}{2} (G_H - G_{OH}) .$$

The observed yields under gamma irradiation agree well with those expected from the known values of the radical and molecular yields from water.

According to the above, the chemistry of  $HO_2$  can be studied by the use of TNM, which readily reacts with  $HO_2$  to form the easily detectable nitroform. This indicator has been used at BNL to determine that  $HO_2$  is actually an intermediate in the familiar reaction of ceric salts with hydrogen peroxide, an assumption often made but never previously verified. TNM does not react when mixed with  $H_2O_2$  or with ceric or cerous salts. When  $Ce^{IV}$ ,  $H_2O_2$ , and TNM solutions are mixed together, the ceric salt immediately reacts with the  $H_2O_2$ , but in addition some TNM is reduced to nitroform, as expected if  $HO_2$  is an intermediate in the reaction. We have found that the consumption of the reagents under different conditions varies just as expected from the reaction sequence:



These reactions occur very rapidly. A complication is that a slower reaction may succeed the above if  $H_2O_2$  is present in excess, which may be expressed by the following sequence:



The rate-determining step here is the formation of pernitrous acid,  $HOONO$ , from nitrous acid and  $H_2O_2$ . The rate of this reaction is well known, and the appearance of nitroform in the mixture, after the initial burst, proceeds at exactly the rate expected from the known kinetics of pernitrous acid formation.

In the decomposition of pure water at high radiation intensities the free radicals  $H$ ,  $OH$ , and  $HO_2$  are formed and combine with one another to produce various reaction products. The reaction products quickly build up to steady-state concentrations which should be proportional to the square root of the radiation intensity. The study of this

system is fraught with experimental difficulties. To obtain the required high intensity, particle-beam sources must be used, but these produce reasonably uniform intensity only over very small volumes. In addition, trace amounts of impurities will falsify the results. By irradiating very small volumes of water and using microanalytical techniques it has been shown that the products do build up to steady-state concentrations which are accurately proportional to the square root of the intensity. The steady state depends on the ratio between the rates of the reactions in which two radicals combine to produce the product and in which a radical reacts with the product to destroy it. It is possible by using a pulsed electron beam to determine the lifetime of the free radicals and hence their absolute reaction rate constants. When the pulse period is short compared to the radical lifetime, the system acts as though the beam were continuous, but of an intensity equal only to the long-time average. When the pulse period is long compared to the radical lifetime, the system acts as though the radiation intensity is that occurring during the individual pulses. In photochemistry this is known as the "rotating sector method" because a light beam is conveniently pulsed by rotating in front of the light source a disk from which a

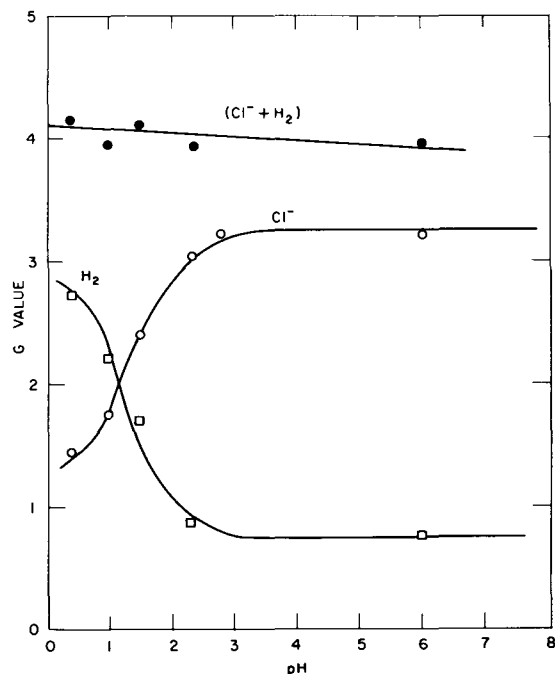


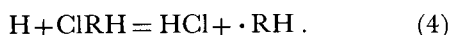
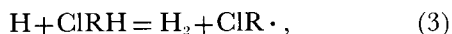
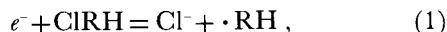
Figure 1. Yield of chloride ion and hydrogen gas from gamma irradiation of chloroacetic acid as a function of pH.

sector has been removed. In the irradiation of water with an electron beam, Ghormley, at Oak Ridge, attempted to modulate the beam by using an actual rotating sector of aluminum. The experiment failed to give accurate results, probably because the scatter of electrons at the edge of the sector prevented the cutoff from light to dark from being sufficiently sharp. With the cooperation of the Instrumentation Division, a device was designed which modulates the electron beam of the Van de Graaff machine electronically. Much difficulty was encountered in installing this device, but a successful design was eventually obtained. A series of pulsed experiments were performed which can be interpreted in terms of the absolute rates of reactions of the OH radical. The absolute rate of the reaction  $\text{OH} + \text{H}_2 = \text{H}_2\text{O} + \text{H}$  is found to be  $4 \times 10^7$  liters/mole-sec at  $23^\circ$ , and for the reaction  $\text{OH} + \text{OH} = \text{H}_2\text{O}_2$  the value is  $8 \times 10^9$  liters/mole-sec.

To determine the lifetime of the  $\text{HO}_2$  radical a different system was set up. This radical is much longer-lived than OH, and it was sufficient to flow a stream of water or solution through the electron beam into a flask containing some substance that would react with  $\text{HO}_2$ . The amount of chemical reaction occurring in the flask depends upon the length of time required for the water to pass from the electron beam to the flask, and hence the decay of  $\text{HO}_2$  can be followed. Most of the determinations have used ferrous sulfate as a reagent; 2,5-dimethyl-*p*-hydroquinone and TNM have also been used. The radicals decay by the expected second-order reaction,  $2\text{HO}_2 = \text{H}_2\text{O}_2 + \text{O}_2$ , but the reaction rate has been found to vary in an unexpected manner with the acidity of the solution, being a minimum at pH 1.8. At pH levels  $>4$ , no  $\text{HO}_2$  could be detected by this method. The rate constant at pH 1.8 is  $\approx 8 \times 10^4$  liters/mole-sec.

It had previously been shown in this Laboratory that the reducing agent produced in water, generally called H, is different from the H atom produced by the free radical oxidation of  $\text{H}_2$  under the same conditions. The latter form, designated  $\text{H}'$ , reacts more slowly with  $\text{H}_2\text{O}_2$ .  $\text{H}_2\text{O}_2$  is also not readily reduced under irradiation in acid solutions, and it appeared that the original H was converted by acid to the other form. This is consistent with the view that the entity formed in the irradiation of water is really a solvated electron rather than an H atom, and that it reacts with  $\text{H}^+$  ions to form H atoms, which are less reactive. A study of

the conversion of these forms has now been carried out with chlorinated organic compounds as indicators. In neutral solutions of chloroacetic acid a prominent product is chloride ion, but with increasing acidity the chloride ion is largely replaced by hydrogen gas as a reaction product. The yields of these products from 0.1 *M* chloroacetic acid solutions adjusted to various acidities with  $\text{H}_2\text{SO}_4$  or  $\text{NaOH}$  are shown as a function of *pH* in Figure 1. The results are consistent with the reaction scheme:



At constant hydrogen ion concentration, the yields of chloride and hydrogen change with increasing concentration of chloroacetic acid, as shown in Figure 2. A change occurs when the concentration of chloro-compound sufficiently exceeds the concentration of hydrogen ion to compete with the  $\text{H}^+$  for reaction with electrons. At high solute concentrations the sum of the two yields increases because of scavenging of electrons from the small regions (spurs) of high free radical concentration. The reactions follow the competition kinetics expected

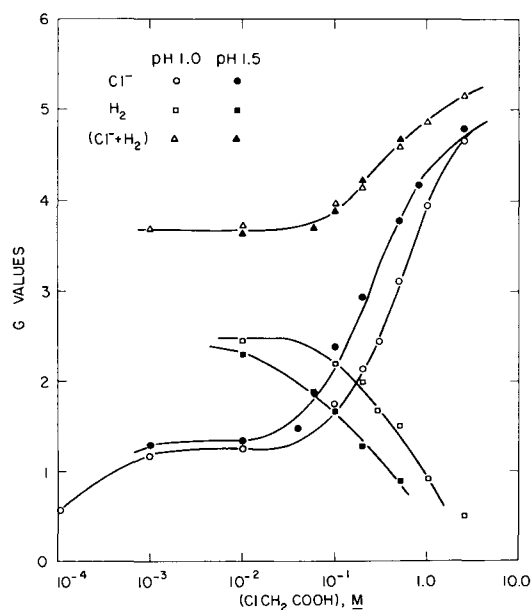


Figure 2. Yield of chloride ion and hydrogen gas from gamma irradiation of chloroacetic acid as a function of acid concentration.

from the above scheme very accurately; the ratio of the rate constants for reactions (2) and (1) is 3.4, and for (4) and (3) is 0.55. A number of other organic chlorine compounds were studied in aqueous solution, including *sym*-dichloroethane, *sym*-dichloroacetone, and  $\alpha$ - and  $\beta$ -chloropropionic acid. All give yields of chloride under gamma radiation that resemble closely the results with chloroacetic acid; some of the compounds (Figure 3) give almost exactly the same results as chloroacetic acid. The effect is not a property of any specific material; it depends only on the nature of the radiolysis of water and of the C—Cl bond. This is the most convincing demonstration yet given for the existence of two types of reducing species in irradiated water. A determination of the true nature of the species will require a great deal of further experimental and theoretical work.

### Organic Compounds

The radiolysis of acetamide ( $\text{CH}_3\text{CONH}_2$ ) was chosen for study because it is a polar organic compound (a class in which little radiation-chemical work has been done), because the amide bond is of particular interest in connection with radiation effects on proteins, and because the material, readily handled in either crystalline or liquid form, is suitable for study of phase effects. The decomposition by gamma-rays is found to proceed by two modes: one yielding  $\text{H}_2$ ,  $\text{CH}_4$ ,  $\text{CO}$ ,  $\text{NH}_3$ , and other products; the second, a split into water and acetonitrile,  $\text{CH}_3\text{CN}$ . The first mode shows yields which are higher in the liquid than in the solid and increase with increasing temperature in either phase. The yield of the other mode, on the contrary, decreases with increasing temperature and is lower in the liquid than in the solid. It is proposed that the formation of the nitrile involves two molecules of acetamide, connected by hydrogen bonds. Following ionization of one molecule, a shift of electrons around the hydrogen-bonded system (of a type familiar in organic chemical reaction mechanisms) can be postulated to lead to the formation of two molecules of nitrile. The reaction naturally becomes less prominent as the bond between the acetamide molecules is weakened with increasing temperature or upon melting. The other mode of decomposition is assumed to be the more usual type of radiation-chemical decomposition involving free radicals.

The radiolysis of *unsaturated* hydrocarbons appears to proceed in part by a direct addition of two

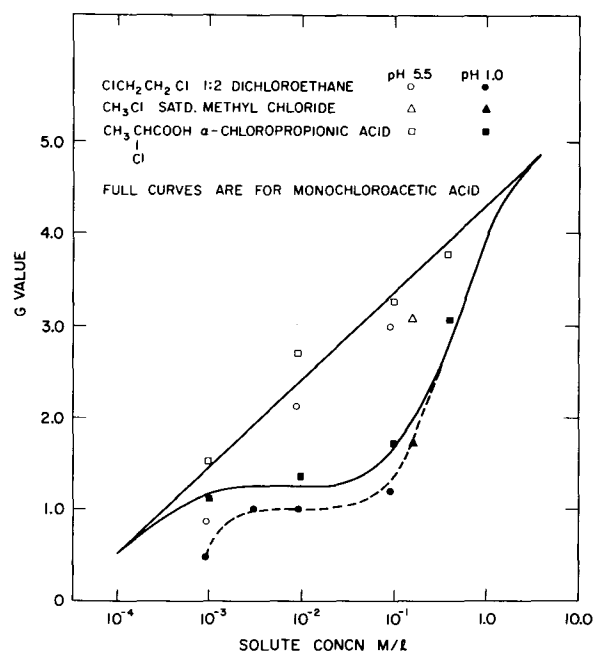


Figure 3. Yield of chloride ion from gamma irradiation of halogenated organic solutes as a function of solute concentration.

molecules at the position of the double bonds. This has been thought to arise from reaction between an ionized molecule of the olefin and a neighboring molecule. If the initially ionized molecule is neutralized before undergoing any chemical reaction, an expected result would be formation of allyl radicals and of hydrogen atoms which would add to neighboring molecules to form alkyl radicals. Table 1 shows some results obtained in the radiolysis of liquid butene-1 and butene-2. The *cis* and *trans* forms of butene-2 were found to give exactly the same results. The allyl radical mechanism would be expected to yield considerable

amounts of diolefin,  $C_8H_{14}$ , in the dimeric fraction of the products (from the combination of two allyl radicals), while the ionic mechanism should give almost entirely mono-olefin,  $C_8H_{16}$ . The results in Table 1 indicate that the mechanism of dimerization of butene-1 may be largely ion-molecule in character, while in butene-2 the allyl-radical mechanism is more important.

A current problem in the radiolysis of *saturated* hydrocarbons is that of whether the breaking of C—H bonds results in the formation of large numbers of free H atoms in the liquid, or whether molecules are brought into reaction with one another by some other means. The mechanism may be studied by addition of various types of material to the hydrocarbon prior to irradiation. At present the radiolysis of pentane to which small quantities of tetrachloroethylene are added is being studied. This material will not add free alkyl radicals and is believed to be inert to free H atoms also, but it should readily react with free electrons. It is found that a few percent of  $C_2Cl_4$  in pentane notably depresses the yield of all products in the pentane radiolysis, but in addition the tetrachloride disappears in quantities considerably greater than the amount by which the pentane radiolysis is depressed. HCl appears in quantities nearly equal to the disappearance of  $C_2Cl_4$ . The results indicate transfer of energy from the pentane to the  $C_2Cl_4$  by some mechanism other than a free-radical or atom reaction, probably by electron capture.

## CHEMICAL EFFECTS OF NUCLEAR TRANSFORMATIONS

### Carbon-14 Recoil Chemistry

The continuing program on the chemistry of recoiling carbon-14 has turned to the investigation

Table 1  
Radiolysis of Butenes in the Liquid Phase With Gamma-Rays

Starting material	$G_{H_2}$	$G$ (double bond isomerization)	$G_{C_2H_4}$	$G$	Extent of unsaturation						Dimeric fraction		
					% $C_8H_{18}$	% $C_8H_{16}$	% $C_8H_{14}$	Carbon skeleton			C—C—C—C	C—C—C—C	C—C—C—C
								C—C—C—C	C—C—C—C	C—C—C—C			
$CH_3CH_2CH=CH_2$	0.6	0.2–0.5	16	1.8	3	94	3	50	23	22			
$CH_3CH=CH-CH_3$ ( <i>cis</i> or <i>trans</i> )	0.7	0.2–0.6	7	1.3	7–8	55	38	28	40	25			

of gaseous systems. Anhydrous ammonia was chosen for this study. Irradiation of ammonia by neutrons produces a uniquely high and specific yield of methane- $C^{14}$ . This high yield of methane- $C^{14}$ , essentially the sole significant product from the operations of the recoil carbon in gaseous ammonia, was not affected by the addition of inert moderating gases. The presence of oxygen as a radical scavenger did not produce any significant effect. When ammonia was used in admixture with methane or methylamine, however, a complex mixture of radioactive products was obtained. The results are summarized in Table 2. Much work remains to be done before this system can be clearly understood. For example, it may be argued that consideration of energy loss probabilities demands a much higher inert moderator-to-reactant ratio in order to achieve efficient cooling of the recoil carbon. If ammonia were responsible for the major portion of the moderation in the mixtures used, then the chemistry of the fragment could not be expected to be very much different from that in

pure ammonia. The effect of the extraneous reactor radiation on product yield and composition has not been determined in this study.

A hypothesis has been suggested which involves hydrogen transfer during a limited but finite lifetime of an intermediate formed by an inelastic collision between the carbon fragment and the hydrogen source. The redistribution of energy and the chemistry possible in the intermediate formed after collision (i.e., a collision complex) appear more important than the absolute amount of translational energy of the reactive fragment just prior to collision.

#### Phosphorus-32 Recoils From Trimethylphosphine

In a number of investigations on  $P^{32}$  recoils from thermal neutron capture in solid and liquid compounds of phosphorus, a considerable fraction of the  $P^{32}$  activity was isolated in the chemical form of the starting material. The suggestion has been made that bond rupture is incomplete. Experi-

Table 2  
Yields of Radioactive Species as a Result  
of the  $N^{14}(n,p)C^{14}$  Nuclear Transformation in Gaseous Ammonia and Methylamine

Material irradiated	Ammonia pressure, atm	Mole fraction of diluent	Ratio of diluent to ammonia	Total $C^{14}$ produced, $\mu C$	Percent distribution of carbon-14*		
					Methane	Methylamine	Others
Ammonia	1	—	—	1.6	89.4	0.10	†
	4	—	—	5.6	104	0.03	†
	8	—	—	10.9	92.1	0.06	†
	12	—	—	14.3	94.4	—	†
Ammonia + neon	8	0.189	0.23	10.0	96.1	0.15	†
	4	0.286	0.41	5.5	96.0	0.14	†
	4	0.475	0.91	5.0	98.0	0.10	†
	1	0.773	3.36	1.33	93.2	0.13	†
Ammonia + xenon	8	0.442	0.79	10.6	102	0.07	†
	4	0.628	1.70	5.3	105	0.06	†
Ammonia + oxygen	4	0.03	0.03	5.8	90	<1	CO <sub>2</sub> 6
							CO ≈0
Ammonia + methane	4	0.667	2.00	5.3	60	≈0.6	C <sub>2</sub> H <sub>6</sub> 6
							Polymer 34
Methylamine (4 atm)	—	—	—	2.79	6.8	1.5	C <sub>2</sub> H <sub>6</sub> 4.5
							CH <sub>3</sub> CN 0.6
							Polymer (l) 10-20

\*The standard deviation of the radiochemical analyses as done by the carrier method is ≈5%. Somewhat larger errors may be involved in values determined by the gas chromatographic flow counting method.

†No other species were detectable by the methods used in these experiments. In addition, HCN was not detectable in neat ammonia.

ments in the condensed phase cannot easily provide information on whether the fraction of product activity in the form of the starting material is due to "inherent retention" or to recombination reactions. In the gas phase recombination reactions can be reduced, and it is possible to test for true retention.

In the present work, gaseous trimethylphosphine was chosen because of its radiation stability and the ease of separating it from phosphine and the other methylphosphines by gas chromatography. Samples of trimethylphosphine were sealed in quartz ampoules in the absence of air and were irradiated under thermal column conditions and in high flux water-cooled facilities. Measurements were made at various initial pressures of phosphine. The gaseous radioactive products found were  $\text{PH}_3$  (main product),  $\text{MePH}_2$ ,  $\text{Me}_2\text{PH}$ , and  $\text{Me}_3\text{P}$ . At 155 mm pressure these contained 21% of the  $\text{P}^{32}$  activity, about 3% being  $\text{Me}_3\text{P}$ . The remainder was found on the walls. At 1.8 mm pressure the yields of all gaseous radioactive products were sharply reduced. This trend would be expected if the inherent retention is zero and the gaseous products, including the parent  $\text{Me}_3\text{P}$ , are formed by recombination reactions. However, it could also be explained if at higher pressure there is inherent retention in the sense that the hot molecule may lose its excess energy by collision with other molecules before it decomposes. To decide between these possibilities, experiments were made at *ca* 1 mm pressure of phosphine, with 50 mm of argon added as a moderator. Over 99% of the activity was found on the walls, and 0.1% or less as  $\text{Me}_3\text{P}$ . The conclusion is that bond-breaking is complete. The effect of argon also indicates that the volatile products are formed while the recoil still has an energy well above thermal.

#### Phosphorus-32 Recoil Chemistry in Inorganic Phosphorus Salts

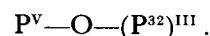
The chemical behavior of  $\text{P}^{32}$  recoil atoms following neutron irradiation of crystals of inorganic phosphorus compounds has been extensively investigated. Further refinement of the technique of paper electrophoresis has permitted the separation, determination, and in a number of cases the identification of the various species present in aqueous solutions of the irradiated compounds. In all, about 22 compounds have been studied. Marked effects of the radiation and thermal environment of the crystals, both during and after

neutron bombardment, were observed. The results obtained differ radically from those of some previous investigators, and it is believed that this difference arises mainly from these environmental effects.

A typical electrophoresis histogram (radioactivity vs distance of movement) is shown in Figure 4. The compound irradiated was anhydrous  $\text{Na}_2\text{HPO}_4$ . The peaks have been tentatively identified, and their percentages determined, as follows:

A	Triphosphosphate	12.2
B	Pyrophosphate	11.7
C	Unknown, probably two species	12.0 total
D	Isohypophosphate	27.1
E	Hypophosphate	2.8
F	Orthophosphate	9.3
G	Diphosphite and possibly another species	7.7 total
H	Phosphite	8.2
I	Hypophosphite	6.8

One of the most interesting results of this investigation is the discovery that the isohypophosphate formed by recoil is uniquely labeled in the trivalent phosphorus position, i.e., it has the structure



#### Recoils in Cobalt Complexes

The technique of paper electrophoresis has also been employed in the study of hot-atom reactions

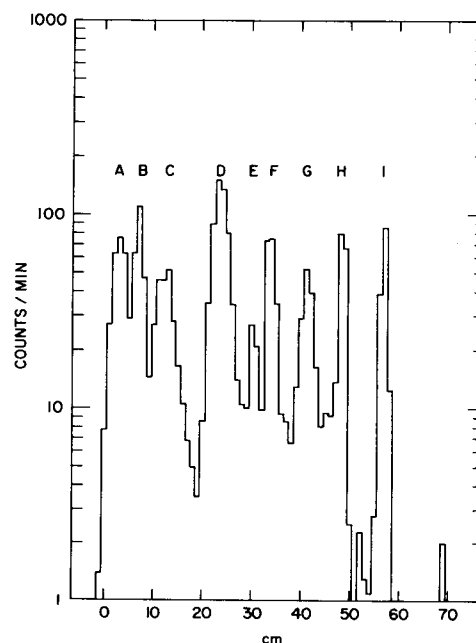


Figure 4. Histogram from paper electrophoresis of neutron-irradiated  $\text{Na}_2\text{HPO}_4$ . Radioactivity is plotted as a function of distance of movement along the paper strip.

in a series of cobalt complexes. It has been found possible to separate a large number of  $\text{Co}^{60}$ -labeled products from irradiated cobaltic hexamine nitrate  $[\text{Co}(\text{NH}_3)_6(\text{NO}_3)_3]$ . Percentages of the different species found after dissolution of crystals irradiated at dry-ice temperature are as follows:

$\text{Co}(\text{NH}_3)_6^{+++}$	6.9
$\text{Co}(\text{NH}_3)_5\text{H}_2\text{O}^{+++}$	4.6
$\text{Co}(\text{NH}_3)_4(\text{H}_2\text{O})_2^{+++}$	0.9
$\text{Co}(\text{NH}_3)_5\text{NO}_2^{++}$	6.7
$\text{Co}(\text{NH}_3)_5\text{NO}_3^{++}$	7.7
$\text{Co}^{++}$	51.4
Unknown No. 1, possibly $\text{Co}(\text{NH}_3)_4(\text{NO}_3)_2^+$	6.9
Unknown No. 2, probably $\text{Co}(\text{NH}_3)_4(\text{NO}_2)_2^+$	ca 8

The balance consists of minor peaks and unresolved activity. Upon annealing for as little as one day at room temperature, the percentage of  $\text{Co}(\text{NH}_3)_6^{+++}$  increases to 25.5, the cobaltous ion correspondingly decreasing. Other complex salts which have been studied include those of nitropentamine, nitratopentamine, and bromopentamine cobaltic ions. In the last case, recoil and annealing reactions of  $\text{Br}^{80m}$ ,  $\text{Br}^{82}$ , and  $\text{Co}^{60}$  have all been investigated in the same crystal; substantial differences are found in the behavior of  $\text{Br}^{80m}$  and  $\text{Br}^{82}$ .

## ORGANIC REACTIONS

### Aromatic Hydrogen Exchange

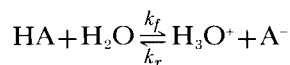
A year ago it was found that aromatic hydrogen exchange in 1,3,5-trimethoxybenzene is subject to general acid catalysis and that this exchange is, therefore, a slow proton transfer reaction ( $A-S_E2$  mechanism). Research has continued on the general features of this first authentic example of an important reaction type. Three areas were investigated: conformance to the Brønsted relation, the form of the dependence of exchange rate on acidity in concentrated aqueous acids, and isotope effects.

Reactions subject to general acid catalysis usually obey a relationship between rate constant ( $k_A$ ) and acid ionization constant of the catalyzing acid ( $K_A$ ) first proposed by Brønsted in 1924:

$$k_A = G(K_A)^\alpha$$

The degree of conformity of hydrogen exchange in trimethoxybenzene to this relationship was tested by measuring exchange rates with seven acids differing in strength over the complete range from  $\text{H}_3\text{O}^+$  to  $\text{H}_2\text{O}$ . Figure 5 shows that the Brønsted relation is obeyed very well; in fact, the present

case is probably one of the best examples of this relation known. This evidence implies that the exchange process must be very similar to the process involved in the ionization reaction



with the equilibrium constant  $K = k_f/k_r$ . It is known that, for a wide variety of acids,  $k_r$  is essentially the rate for encounter of  $\text{H}_3\text{O}^+$  and  $\text{A}^-$  ions. The observed range of values for  $K$  must result from the variation in  $k_f$ , the rate constant for proton transfer from acid to substrate. Similarly, a slow proton transfer must be involved in the hydrogen exchange of trimethoxybenzene, since the Brønsted relationship is obeyed.

Rate measurements of aromatic hydrogen exchange of trimethoxybenzene with aqueous perchloric acid up to a concentration of 3 *M* showed that this reaction depends on the Hammett acidity function  $H_0$ . This acidity dependence is midway between two others predicted on the basis of models for the reaction involving no transfer and complete transfer of the proton. Thus, the results indicate that the proton is, in fact, in the process of being transferred at the transition state.

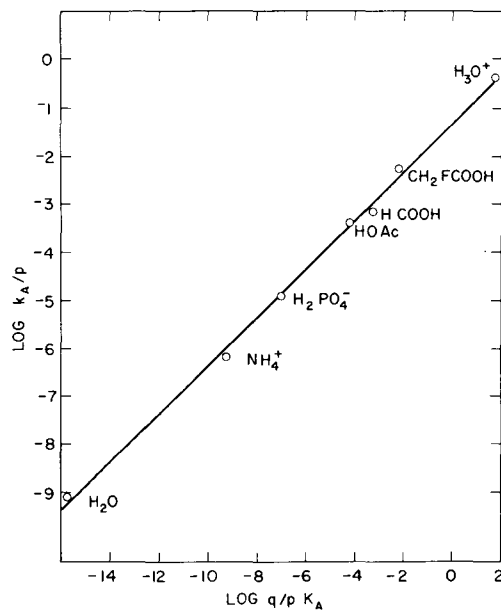


Figure 5. Logarithm of the rate constant for the acid-catalyzed hydrogen exchange of trimethoxybenzene plotted against logarithm of the ionization constant of the catalyzing acid.



From a combination of several experiments involving all three hydrogen isotopes, it is possible to obtain the deuterium isotope effect for proton loss from the hydronium ion and from the protonated trimethoxybenzene intermediate. The value of 7 found for the latter is in the normal range for a deuterium isotope effect. The unusually low factor for the relative rates of proton and deuterium transfer from the hydronium ion presumably arises because the loss of a proton from  $\text{H}_3\text{O}^+$  produces a water molecule with stronger O—H bonds than those in the hydronium ion.

### Secondary Isotope Effects

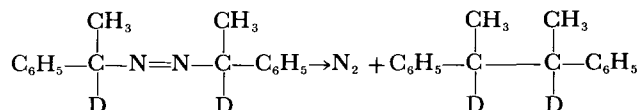
The secondary  $\alpha$ -deuterium isotope effect in the *cis-trans* isomerization of maleic acid-2,3- $d_2$  catalyzed by aqueous potassium thiocyanate has been measured. The mechanism of this reaction, originally postulated by Nozaki and Ogg, involves a rapid pre-equilibrium step in which a carbonyl-oxygen is protonated. Isomerization, by rapid rotation about the central carbon-carbon bond, occurs after the slow approach of thiocyanate ion to the ethylenic-carbon atom. The final conversion of the complex of fumaric acid proceeds by loss of thiocyanate ion followed by removal of the proton. The observed inverse isotope effect ( $k_{\text{H}}/k_{\text{D}} = 0.88$  at  $80^\circ\text{C}$ ) is consistent with Streitwieser's explanation for this type of secondary isotope effect. The main effect produced by a change from a tetrahedral,  $sp^3$ -hybridized carbon-hydrogen bond to a trigonal,  $sp^2$ -hybridized bond is a decrease in the carbon-hydrogen out-of-plane bending frequency. This change from tetrahedral to trigonal configuration produces a normal isotope effect. In the maleic acid isomerization, however, the change is in the opposite direction and an inverse isotope effect is therefore expected. The observed inverse isotope effect is consistent with the postulated mechanism.

This isotope effect has also been measured as a function of temperature. By applying some simplifying assumptions to the theory of isotopic rate constants, it is found that a plot of  $\log k_{\text{H}}/k_{\text{D}}$  against the reciprocal of the absolute temperature yields the sum of the differences in frequencies between the ground and transition states. The result obtained is about  $375\text{ cm}^{-1}$ .

The infrared spectra of maleic anhydride (a model for maleic acid) and maleic anhydride- $d_2$  were examined to determine carbon-hydrogen bending frequencies. These frequencies, combined

with those for tetrahedral C—H, give a difference of about  $565\text{ cm}^{-1}$  for the complete change from trigonal to tetrahedral carbon. Therefore, it might be said that the transition state is slightly more than one-half of the way from an olefinic ground state to a tetrahedral product.

The secondary  $\alpha$ -deuterium isotope effect has also been measured for the free radical decomposition of azobis- $\alpha$ -phenylethane- $\alpha,\alpha'$ - $d_2$  in ethylbenzene at  $105^\circ\text{C}$ . In this reaction,  $k_{\text{H}}/k_{\text{D}} = 1.25$  at  $110^\circ\text{C}$ , where the normal molecule is compared

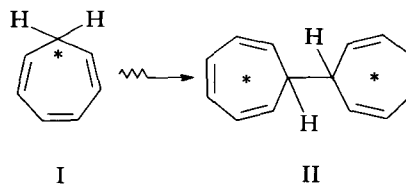


with one containing two deuterium atoms. The few available data on secondary  $\alpha$ -deuterium isotope effects give  $k_{\text{H}}/k_{\text{D}} \approx 1.15$  at about  $60^\circ\text{C}$ , and this ratio can be extrapolated to about 1.12 at  $105^\circ\text{C}$ . Since the effect observed in the free radical decomposition above is twice this large, it appears that both carbon-nitrogen bonds are simultaneously breaking in the transition state.

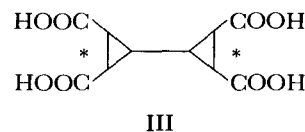
### Irradiation of Cycloheptatriene-1- $\text{C}^{14}$

The radiation chemistry of compounds labeled with carbon-14 affords a sensitive method for determining yields and for studying skeletal rearrangement. Very little has been done in this respect in the field of radiation chemistry.

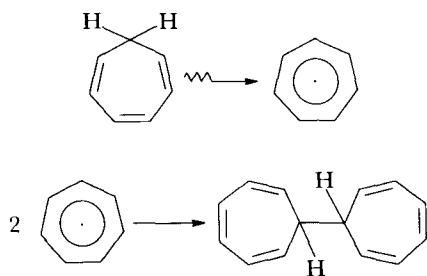
The irradiation of cycloheptatriene-1- $\text{C}^{14}$  (I) in a nuclear reactor or in a beam of 1.95-Mev electrons from a Van de Graaff generator leads to the formation of labeled ditropyl (II), among other prod-



ucts. Sensible amounts of labeled ditropyl were isolated by carrier techniques, and, by the peroxide cleavage of the ozonide, were converted to dicyclopropylyltetracarboxylic acid (III):



The point studied was whether or not the reaction proceeds in a way that distributes the  $C^{14}$  randomly in the final carbon skeleton. Complete randomization would allow only five-sevenths of the activity to appear in the acid (III), while nonrandomization of activity would require that 100% of the activity, originally in the 1 position, be accounted for by the acid (III). The activity found in the acid (III) was, in fact, five-sevenths of that originally located in the ditropyl. This result suggests that a symmetrical precursor is involved in ditropyl formation, e.g., according to the following scheme involving a resonance stabilized tropylium radical:



### Biosynthesis of Nicotine

Sterile cultures of certain tobacco roots have been shown previously to produce nicotine which incorporates, without scrambling of the label, nicotinic acid labeled in the 2, 4, and 5 positions. However, when nicotinic acid-6- $t$  is supplied, almost no tritium appears in the nicotine produced. Since it seemed likely that the mode of incorporation of nicotinic acid was via oxidative attack at the 6 position, 6-hydroxynicotinic acid- $N^{15}$  and 1-methyl-6-oxonicotinamide-2- $t$  were supplied to root cultures, but in these cases no label appeared in the nicotine produced. The possibility that incorporation proceeds through a 1,6-dihydronicotinic acid derivative is now being considered.

It has also been shown that nicotinic acid is not produced by the tobacco plants from the usual precursors of this substance, such as anthranilic acid, tryptophan, or kynurenine. A study is now being made to determine an alternate pathway which can be utilized by tobacco roots for the synthesis of nicotinic acid, and to this end a number of small molecules, labeled in various positions, have been supplied to the root cultures. Very substantial incorporation occurred with  $\beta$ -alanine-2- $C^{14}$ , but not with the 1- or 3-labeled material. Substantial (but less) labeling was found with

similarly labeled fumaric, aspartic, malonic, and succinic acids, acetate-2- $C^{14}$ , glycerol-1,3- $C^{14}$ , glucose-6- $C^{14}$ , and uniformly labeled alanine (alanine-1- $C^{14}$  is not incorporated). The distribution of activity between the two rings of the nicotine molecule has been determined for these cases, and some hints of ways in which these compounds may be utilized for the synthesis of the pyridine ring are beginning to emerge. At this point it appears likely that at least some of these compounds are being converted to acetate through involvement in the Krebs citric acid cycle before being incorporated into nicotinic acid by the tobacco roots. Some, however, may be found to be incorporated by more direct means into certain parts of the pyridine ring.

### NUCLEAR CHEMISTRY

A number of decay scheme studies discussed in last year's report have been completed during the past year. Among these are the investigations of the decay of  $Zn^{63}$ , of the assignment and properties of several gadolinium and europium isotopes, and of the creation of double  $K$ -vacancies in the  $K$ -electron capture of  $Cs^{131}$ . The study of resonance fluorescence of  $\gamma$ -rays in  $Cu^{63}$  undertaken in collaboration with members of the Physics Department has also been carried to successful conclusion and is discussed in the Physics section of this report.

Assembly of a 50-cm radius, double-focusing  $\beta$ -ray spectrometer, under way since August 1959, has been completed, and performance measurements and adjustments are now being made. Early results indicate that the instrument can achieve resolutions of  $\approx 0.1\%$  with transmission of  $\approx 0.05\%$ .

The main effort in nuclear chemistry in the past year has again been spent in the study of nuclear reactions. With the resumption of Cosmotron operation during the year and with the prospect of the AGS start-up, emphasis has returned to high energy reactions, although considerable work is still being done on low energy reactions, primarily with the 60-in. cyclotron.

### High Energy Nuclear Reactions

The techniques used in nuclear reaction studies have included, besides the usual radiochemical determinations of the yields and properties of reaction products, measurements of reaction yields

by mass spectrometry and nuclear emulsion detection. In addition, a start has been made towards adapting solid state detectors of the new junction type for use in nuclear reaction work; initial tests were most encouraging and showed that these devices, which have much better energy resolution than do scintillation counters, can be used successfully for detection and measurement not only of protons, deuterons, and  $\alpha$ -particles of energies up to 10 to 40 Mev, but also of  $\beta$ -particles, low energy  $\gamma$ -rays, and minimum ionizing particles.

Nuclear emulsion studies have continued on the interactions of high energy protons with silver and bromine nuclei. It was observed that with 1.0-Bev protons  $\approx 3\%$  of the interactions lead to fission. At 2.0 and 3.0 Bev this increases to  $\approx 7\%$  and  $\approx 11\%$ , respectively. Although the distribution of products is very wide, symmetric fission is most probable; in about two-thirds of the fission events the ratio of track lengths is between 1 and 2. In most of the fission events,  $\alpha$ -particles and/or other light particles ( $Z \leq 6$ ) were also observed. Protons were not recorded in the emulsion used. From measurements of the angular distributions of the  $\alpha$ - and other light particles with respect to the fission fragments, conclusions could be drawn about the time sequence of fission and particle emission. Emission of particles simultaneous with fission would lead to a peak near  $90^\circ$ , while emission of the particles subsequent to fission would result in forward-backward peaking. The experiments showed a nearly isotropic angular distribution of the  $\alpha$ -particles (and light particles) with respect to the fission fragments. This indicates that particle emission precedes fission in most cases.

Considerable success has been achieved in accounting for the yields of very light nuclides such as  $\text{He}^6$ ,  $\text{Be}^7$ , and  $\text{Li}^8$  by an evaporation mechanism in which they, along with neutrons, protons, and  $\alpha$ -particles, are boiled out of the highly excited nuclei resulting from the interaction of high energy protons with heavy target atoms. To determine how far this approach may be extended, it is necessary to compare observed formation cross sections with theoretical predictions for the yields of heavier evaporated particles. One difficulty in calculating the evaporation probability of a particle is that the emission of the particle must be computed not only in its ground state but also in any of its bound excited states, with appropriate statistical weights given to each state. Therefore all the bound excited states of the particle and

their spins must be known. Since these data are often uncertain, the calculations are also open to question. As far as ease of calculation is concerned,  $\text{N}^{13}$  with no bound excited states is the obvious choice for comparison between experiment and theory.

The experimental work on this problem has been completed, and cross sections for  $\text{N}^{13}$  formation in aluminum, zinc, indium, lead, and uranium targets have been measured with protons of 1, 2, and 3 Bev. The formation cross sections all rise rapidly with energy; at all energies studied the yields are highest from aluminum and decrease regularly as heavier targets are considered. Evaporation calculations in these systems are now being performed at the Weizmann Institute and preliminary results show at least rough agreement with the experimental numbers, which indicates that an evaporation mechanism may indeed be largely responsible for  $\text{N}^{13}$  formation.

Study of the recoils from the high energy reaction  $\text{Al}^{27}(p, 3pn)\text{Na}^{24}$  has been continuing. A method has been devised for analyzing the angular distribution data obtained several years ago. The thick target range studies have been extended up to 3 Bev. All the recoil properties studied so far are consistent with the following model of the mechanism of the reaction. The high energy proton undergoes one glancing collision with a nucleon in the nucleus and escapes. The low energy struck nucleon is captured; it transfers its momentum to the nucleus and deposits its kinetic energy in the nucleus in the form of excitation. Those cascades leading to  $\text{Na}^{24}$  have a constant deposition energy of about 55 Mev, which is then used to evaporate the three remaining particles. Further experimental studies of a more detailed nature are in progress to verify this model.

In a more general attempt to gain insight into the expected behavior of recoiling nuclei formed in high energy reactions, the momentum imparted to nuclei in the cascade process has been calculated with the aid of the results of the recent Monte Carlo calculation of intranuclear cascades. Results were obtained for 0.46 to 1.84-Bev protons incident on  $\text{Ru}^{100}$ ,  $\text{Bi}^{209}$ , and  $\text{U}^{238}$ . The forward and transverse components of momentum of the residual nucleus exhibit a wide range of possible values and are, on the average, approximately equal. The average forward component of momentum increases linearly with the excitation energy of the residual nucleus. The relation between

these two quantities leads to considerably lower values of the average excitation energy associated with experimentally determined values of the forward component of momentum than did the relations used previously. The calculated momentum values are in most cases consistent with experimental results.

An attempt to observe the  $\text{Pu}^{239}(p, \pi^-)$  reaction has been made. The measurement of the cross section for this reaction and of the range of the product could provide information on the momentum distribution of neutrons in the target nucleus because the internal motion of the neutrons is probably important for this reaction. The particular choice of target was dictated by a desire to minimize the contribution of the secondary  $(\alpha, 3n)$  reaction to the formation of the measured product and by the high sensitivity available for the detection of the  $\text{Cm}^{240}$  product by use of  $\alpha$  pulse height analysis. Experiments with 380-Mev protons yield a formation cross section of  $7 \times 10^{-6}$  mb for  $\text{Cm}^{240}$ . This value is consistent with the expected cross section for an  $(\alpha, 3n)$  secondary reaction, so that the cross section for the  $(p, \pi^-)$  reaction is even lower. It is planned to repeat this experiment at a bombarding energy only slightly above the reaction threshold.

Studies of uranium fission by protons in the BeV range have continued. The cross sections for formation of a number of calcium, scandium, vanadium, and chromium isotopes are being measured in order to obtain further information on the mass yield curve and isobaric yield distributions in this region. The mass spectrometric study of cesium yields in uranium fission, interrupted by the 1957 breakdown of the Cosmotron, was resumed. It has been supplemented by radiochemical measurements of the formation cross sections of the shorter-lived cesium isotopes. Where both techniques could be used for the same nuclide, the results agree very well. Data have now been collected at a number of proton energies between 100 Mev and 6 BeV, and at most of these energies independent formation cross sections of  $\text{Cs}^{129}$ ,  $\text{Cs}^{130}$ ,  $\text{Cs}^{131}$ ,  $\text{Cs}^{132}$ ,  $\text{Cs}^{134}$ ,  $\text{Cs}^{135}$ , and  $\text{Cs}^{136}$ , as well as chain yields of  $\text{Cs}^{125}$ ,  $\text{Cs}^{127}$ ,  $\text{Cs}^{129}$ ,  $\text{Cs}^{131}$ ,  $\text{Cs}^{135}$ , and  $\text{Cs}^{137}$ , will be available. Although final computation and interpretation of the data have not been completed, it appears that two of the most interesting results will be (a) the low bombarding energies at which some of the neutron-deficient products begin to be formed, and (b) the fact (previously sus-

pected but not firmly established) that at the highest energies studied ( $>2$  BeV) the isobaric yield distributions in this mass region appear to have peaks on both the neutron-excess and neutron-deficient side of stability, with a dip in between.

In addition to the high energy proton work it has for the first time been possible to study radiochemically nuclear reactions induced by high energy pions. Part of a "high intensity" ( $\approx 10^5$  per Cosmotron pulse) beam of 1.6-BeV  $\pi^-$ -mesons was available during a long bubble chamber experiment, and mercury targets of  $\approx 6$  kg were exposed in this beam. A number of reaction products were isolated. This was in the nature of a preliminary experiment to establish that radiochemical studies of pion-induced reactions could be successfully carried out at the intensity levels available. Indications are that the cross section pattern may differ in some significant respects from that found in proton interactions. A plastic scintillation detector with light integrator for monitoring  $\pi$ -beams has now been constructed and tested, and further experiments with high energy  $\pi^-$  irradiations of mercury are planned.

### Low Energy Nuclear Reactions

The use of the 60-in. cyclotron for the study of low energy nuclear reactions to provide information required in the analysis of high energy nuclear reactions has also contributed to an understanding of the relative importance of "compound processes" and "direct processes" in the low energy region. One way of estimating the relative importance of these two processes is to compare the experimental results and those expected from the statistical theory of compound-nucleus decay. The latter may be obtained from a Monte Carlo calculation that was described in a previous report. In this manner, for example, it was found in the past year that the main features of the reactions between  $\text{Sc}^{45}$  nuclei and  $\alpha$ -particles with energies up to 40 Mev could be described rather well within the compound-nucleus formalism. This study included the determination of excitation functions for the  $(\alpha, n)$ ,  $(\alpha, 2n)$ ,  $(\alpha, 2p)$ ,  $(\alpha, 2pn)$ ,  $(\alpha, \alpha n)$  and  $(\alpha, \alpha 2n)$  reactions of  $\text{Sc}^{45}$ . The success of the compound-nucleus theory in predicting the approximate shapes and magnitudes of excitation functions in this and many other instances encouraged extension of the calculation to more sensitive quantities, such as the energy dependence of the

ratio of cross sections for  $(\alpha, p)$  and  $(\alpha, n)$  reactions extending up to energies where more complex reactions are more probable. With the aid of a program written for the IBM 610 computer this ratio was calculated from the statistical theory; with suitable parameter choices, the compound-nucleus model was thus found to be useful even for the estimation of quantities as sensitive as this ratio.

An inviolable requirement of the compound-nucleus theory, independent of detailed assumptions, is that the decay properties of an excited compound nucleus must be independent of its mode of formation. To test the validity of this postulate, an investigation of the reactions of  $\alpha$ -particles with  $\text{Co}^{59}$  and protons with  $\text{Ni}^{62}$  is under way. If compound-nucleus formation takes place, both bombardment systems will lead to a  $\text{Cu}^{63}$  compound nucleus. Preliminary results indicate that the ratio of cross sections for the  $(p, \alpha n)$  reactions to the  $(p, \alpha 2n)$  reaction is the same as that for the  $(\alpha, \alpha n)$  to the  $(\alpha, \alpha 2n)$  reactions when the energies of the protons and  $\alpha$ -particles are such that the excitation energy of the compound nucleus is the same. Thus these particular reactions seem to conform to a fundamental requirement of compound-nucleus theory.

On the other hand, the results obtained in an investigation of the reactions of  $\alpha$ -particles with  $\text{Sn}^{124}$  deviate, even in gross behavior, from those predicted by the usual formulation of compound-nucleus theory. The nature of the deviation [the cross sections for the  $(\alpha, p)$  and  $(\alpha, n)$  reactions do not decrease rapidly enough with increasing energy of the  $\alpha$ -particles] is in the proper direction for a contribution to these reactions from direct processes.

In another attempt to distinguish between compound-nucleus and direct interaction mechanisms, a comparison of the  $(\alpha, \alpha n)$  and  $(\alpha, \alpha p)$  reactions of  $\text{Ce}^{142}$  was undertaken. In the helium ion range from 25 to 41 Mev, the cross section of the  $(\alpha, \alpha n)$  reaction was found to rise monotonically, reaching a value of  $62 \pm 5$  mb at 41 Mev. The  $(\alpha, \alpha p)$  reaction could not be definitely established, but an upper limit of 0.1 mb was set for its cross section at all energies up to 41 Mev. This result appears to rule out a direct knock-on mechanism (which would result in comparable knock-out probabilities for neutrons and protons); however, it makes a compound-nucleus mechanism seem very unlikely also, since evaporation calculations with reasonable parameter choices predict an  $(\alpha, \alpha n)$  excita-

tion function with a maximum somewhere in the energy region studied. The existence of a measurable cross section for the  $\text{Ce}^{142}(\alpha, 2pn)$  reaction in this energy region (2.5 mb at 41 Mev) found as a by-product of this work suggests that this reaction proceeds mainly by  $\text{He}^3$  emission, probably by a stripping mechanism. These results are consistent with the hypothesis that, in this energy range, at most one particle is emitted as a result of direct interaction.

Excitation functions have been measured for the formation of  $\text{Cd}^{111m}$  and  $\text{In}^{115m}$  in the bombardment of  $\text{Cd}^{111}$  and  $\text{In}^{115}$ , respectively, with 6 to 10-Mev protons, 10 to 20-Mev deuterons, and 15 to 40-Mev  $\alpha$ -particles. The excitation function for the  $\text{In}^{115}(\alpha, \alpha n)\text{In}^{114m}$  reaction has also been obtained. All the excitation functions increase monotonically with energy. The cross sections for the formation of  $\text{In}^{115m}$  at the highest bombarding energies are 1.5 mb, 44 mb, and 5.9 mb, for the proton-, deuteron-, and alpha-induced reactions, respectively. The corresponding cross sections for the formation of  $\text{Cd}^{111m}$  are 2 to 3 times larger. The results have been compared with the predictions of the statistical theory. A definite conclusion can be drawn only in the case of the  $(\alpha, \alpha \gamma)$  reaction, where it is shown that an evaporation mechanism cannot account for the measured cross sections at bombarding energies above 25 Mev. The contribution of Coulomb excitation to the observed yields has been calculated. It is found that this process contributes substantially to the  $(\alpha, \alpha \gamma)$  and  $(p, p \gamma)$  reactions, particularly at low bombarding energies.

Most existing estimates of the variations of nuclear level density with excitation energy were made in either of two ways: (1) by the analysis of excitation functions, or (2) by analysis of the energy spectra of emitted particles. The analyses for both types of data are customarily performed assuming the compound-nucleus mechanism. In 1956, Igo and Wegner pointed out that the results of these two approaches are in sharp disagreement, especially for nuclei with  $A > 50$ .

Since the excitation function of a compound-nucleus reaction is probably least distorted by direct interaction contributions nearest the reaction threshold, an attempt is being made to analyze the threshold behavior of excitation functions. Such an analysis cannot be made by employing only the usual formulas for the spectra of emitted particles in Weisskopf's "spinless" approximation. The

spinless formulas are reasonably accurate only if the last particle emitted can still form a product with sufficient excitation energy that many levels with wide distribution of spins can be populated. Near threshold, only one or a few levels are available. Consequently the emission of the last particle competes with other modes of de-excitation (especially  $\gamma$ -ray emission) in a way that requires a separate detailed analysis for each case.

A suitable program of analysis for the threshold behavior of excitation functions has been developed and applied tentatively to two cases: the excitation functions for  $\text{Bi}^{209}(p,2n)\text{Po}^{208}$  (the data of Andre et al.), and  $\text{Sm}^{144}(\alpha,3n)\text{Gd}^{145}$  (work in progress at Brookhaven). Preliminary results indicate that the level density behavior inferred for both cases agrees well with level density information obtained from most analyses of emitted particle spectra.

Calculations have been made of the effects of high angular momentum on the evaporation of neutrons from an excited nucleus and of the effects of such evaporation on the angular momentum of the residual nucleus. Two different assumptions about the density of states have been made: that the density of states goes, in one case, as  $\exp\sqrt{2aE}$ , or, in the other, as  $\exp E/T$ , where  $E$  is the excitation energy and  $a$  and  $T$  are constants. The results show that when the first assumption is used the probability for neutron emission and the average kinetic energy of emitted neutrons both decrease as the angular momentum of the compound nucleus is increased. This behavior is reversed if the second assumption is used. The variation of average kinetic energy with angular momentum agrees well with a simple classical model.

### Spatial Constancy of Cosmic Rays

Since cosmic-ray intensity has been shown to increase with solar flare activity, it is clear that at least a part of cosmic radiation is produced by the sun. However, for various reasons it is now believed that the sun is not the major source of cosmic radiation. Meteorites striking the earth have highly eccentric orbits such that their distance from the sun ranges from  $<1$  to  $>3$  astronomical units, and they have been moving in these orbits for 20 million years or more. Thus, a study of cosmic-ray spallation products in a meteorite should yield information about cosmic rays in other parts of the solar system.

A test of the spatial constancy of cosmic rays may be made by measuring the ratio of a short and a long-lived radioactive spallation product in a recently fallen meteorite. The amount of the isotope with the short half-life reflects the cosmic-ray intensity over the last portion of the meteorite's orbit before it intersected the earth's orbit, while the amount of the isotope with the long half-life gives a measure of the cosmic-ray intensity averaged over the entire orbit. This ratio must then be compared to the production rates of these two isotopes as obtained by bombardment of a sample of the meteorite with high energy protons from an accelerator.

$\text{Ar}^{37}$  with its half-life of 35 days and  $\text{Ar}^{39}$  with its half-life of 325 years are particularly well suited for this purpose. Extraction and purification of these isotopes is relatively easy, and the subsequent counting can be done with counters having a high counting efficiency and very low backgrounds.

Such a measurement was made on the Hamlet chondrite which fell in Indiana on October 13, 1959. The average  $\text{Ar}^{37}/\text{Ar}^{39}$  ratio for two samples of this meteorite was found to be  $2.0 \pm 0.3$ .

To test the spatial constancy of cosmic radiation, the relative production rates of  $\text{Ar}^{37}$  and  $\text{Ar}^{39}$  under a constant cosmic-ray flux must be known.

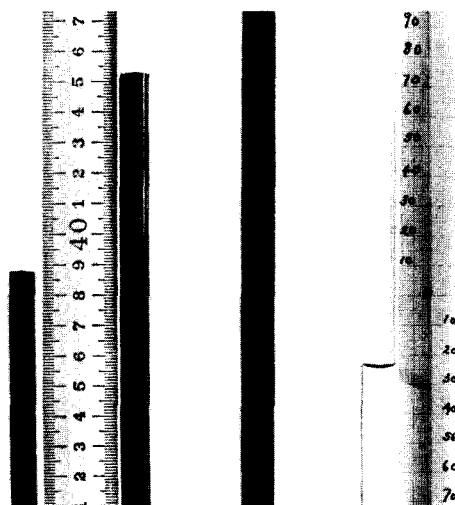


Figure 6. Difference in the vapor pressures of  $\text{Ne}^{20}$  and  $\text{Ne}^{22}$  at  $21.2^\circ\text{K}$ . The mercury manometer on the left registers the absolute vapor pressure of isotopically normal neon. The oil manometer on the right (14 times as sensitive as the mercury manometer) registers the difference in vapor pressures of 99%  $\text{Ne}^{20}$  and 71.2%  $\text{Ne}^{22}$ .

This ratio was approximated by bombarding a third sample of Hamlet with 3-Bev protons from the Cosmotron, and was found to be  $1.5 \pm 0.2$ . Another value for this ratio may be derived by using the individual  $\text{Ar}^{37}/\text{Ar}^{39}$  production ratios for the elements iron, calcium, and potassium normalized for the relative abundance of these elements in the meteorite. These are the only elements in a meteorite with sufficiently high mass number and in sufficient abundance to give appreciable amounts of argon isotopes. The value of  $\text{Ar}^{37}/\text{Ar}^{39}$  so derived was  $1.7 \pm 0.2$ .

By comparing these ratios with the value of  $2.0 \pm 0.3$  obtained on the Hamlet meteorite from cosmic-ray bombardment, it may be concluded that the average cosmic-ray flux over many revolutions of the meteorite in its orbit was the same as the cosmic-ray flux in the vicinity of the earth's orbit, within the limits of error of the experiment. In addition, it may be concluded that the sun is not the main source of cosmic radiation.

## CHEMISTRY OF ISOTOPES

### Isotope Effects on Vapor Pressures

Recently there has been considerable interest in the effect of isotopic substitution on properties of condensed states. In addition to their application to isotope separation, such effects provide a basis for the development of the theory of condensed phases. Atomic solids provide a starting point in such a development, since complications produced by the coupling of inter- and intramolecular motions do not arise.

For this reason, the vapor pressures of  $\text{Ne}^{20}$  and  $\text{Ne}^{22}$  have been measured over the range 16.4 to 30.1° K (Figure 6). Measurements were made with a differential manometer which compared the vapor pressures of 99.9%  $\text{Ne}^{20}$  and 72.2%  $\text{Ne}^{22}$ . The results, shown in Figure 7, can be extrapolated to the pure isotopic species with sufficient accuracy. For solid neon, the data can be accounted for to high accuracy by using an Einstein lattice or a Debye lattice with  $\theta_D(\text{Ne}^{20}) = 74.6^\circ \text{K}$ . This value may be compared with one of  $73^\circ \text{K}$ , which is obtained from neutron scattering intensities and is also independently obtained from a theoretical calculation of the zero-point energy of the solid.

The difference between the  $\text{Ne}^{20}/\text{Ne}^{22}$  vapor pressure ratio for the liquid and the solid can be explained by the change in the number of nearest neighbors surrounding an individual neon atom.

Neutron diffraction data show that this number decreases from 12 in the solid to 8.8 in the liquid, while the average interatomic distance does not change.

For the slightly more complicated case of a diatomic molecule, a theoretical treatment of isotope effects on the vapor pressures of isotopic species of nitric oxide has been carried out. The experimental data for ratios of vapor pressures of  $\text{N}^{15}\text{O}^{18}$ ,  $\text{N}^{15}\text{O}^{16}$ , and  $\text{N}^{14}\text{O}^{18}$  relative to  $\text{N}^{14}\text{O}^{16}$  can be represented by the equation

$$\ln P'/P = A/T^2,$$

which is of the form predicted by the theory of isotope effects when quantum corrections are small. This theory also predicts that the vapor pressure ratio for one such pair of isotopic species (e.g.,  $\text{N}^{15}\text{O}^{18}/\text{N}^{14}\text{O}^{16}$ ) is uniquely determined by the ratios for the other two species ( $\text{N}^{15}\text{O}^{16}/\text{N}^{14}\text{O}^{16}$  and  $\text{N}^{14}\text{O}^{18}/\text{N}^{14}\text{O}^{16}$ ). This is indeed found to be the case. Similar reactions are found for differences in the triple point, heat of vaporization, and boiling point. From a consideration of the probable structure of the dimer, which is known to predominate in solid nitric oxide, it can be predicted

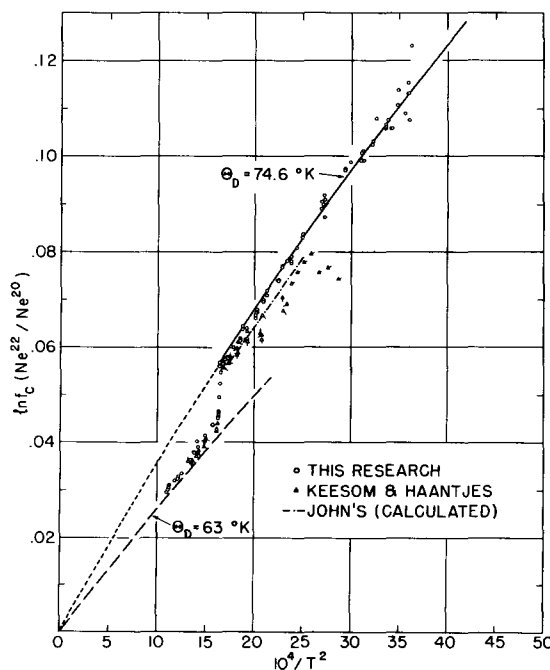


Figure 7. Ratio of vapor pressures of neon isotopes,  $P_{\text{Ne}^{20}}/P_{\text{Ne}^{22}}$ , as a function of the inverse square of the temperature. Data are calculated for an ideal gas in equilibrium with a solid of negligible molar volume.



that  $\ln(P_{N^{14}O^{16}}/P_{N^{14}O^{18}}) = 1.46 \ln(P_{N^{14}O^{16}}/P_{N^{15}O^{16}})$ . The experimental value is 1.38, in satisfactory agreement. The same relative effect of  $N^{15}$  and  $O^{18}$  on the difference in heats of vaporization and triple points is predicted, while the experimental values are 1.45 and 1.40. Finally, from the observed Raman spectrum of liquid NO, the contribution to the vapor pressure ratio from vibrations within the dimer can be evaluated. This contribution, together with a correction of about 20% for dimer-dimer interactions, leads to a value of  $P_{N^{14}O^{16}}/P_{N^{15}O^{16}} = 1.025$  compared with the experimental one of 1.0295. Thus the differences in thermodynamic properties of isotopic nitric oxide molecules can be quantitatively calculated in terms of a structure of weakly interacting dimers.

### Transition Metal Ions in Light and Heavy Water

Recent work here and in other laboratories has shown that spectra and chemical properties of transition metal ions differ in light and heavy water. Since hydrogen bonds are stronger in  $D_2O$  than in  $H_2O$ ,  $D_2O$  is a poorer solvent for neutral solutes than is  $H_2O$ . However, anions are more strongly solvated in  $D_2O$  than in  $H_2O$ , by virtue of hydrogen bonding or charge-dipole interaction between the anion and solvent.

In the case of cation complexes containing water molecules in the inner coordination sphere, there will be a contribution to the vibrational zero-point energy from modes in which the water molecules move with respect to the cation. The predicted zero-point energy difference between  $M(H_2O)_6^{+3}$  and  $M(D_2O)_6^{+3}$  is about  $7500 \text{ cm}^{-1}$ , which would account for observed changes in electronic spectra.

In addition, the interaction of the cation charge with the water molecule dipole will decrease the internal frequencies of a coordinated water molecule compared to the frequencies in the solvent. The isotopic shift in zero-point energies produced by this effect will stabilize a complex of the type  $M(H_2O)_5X^{+2}$  to a greater extent in  $D_2O$  than in  $H_2O$ . These effects may determine relative rates of electron transfer reactions in light and heavy water.

## STRUCTURAL CHEMISTRY

### Neutron Diffraction by Magnetic Crystals

Until very recently, the antiferromagnetic compounds whose magnetic structures have been in-

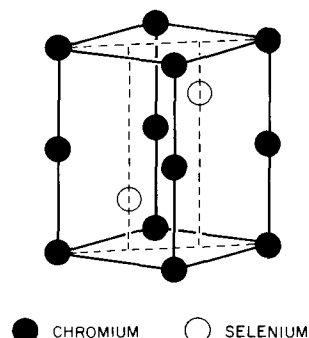


Figure 8. Crystal structure of CrSe.

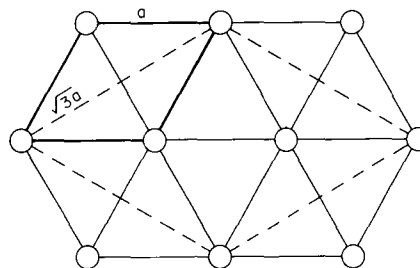


Figure 9. Basal plane enlargement of chemical unit cell of CrSe (solid lines) to magnetic unit cell (broken lines). The  $c$  axis is the same for both.

vestigated by neutron diffraction techniques were all satisfactorily described in terms of a collinear spin arrangement, i.e., a simple  $180^\circ$  reversal of spins. The application of magnetic space groups previously discussed in these reports has pointed up more strongly than ever the possibility of non-collinear spin arrangements. A likely situation for the occurrence of this configuration is in a system with hexagonal symmetry. A large number of the sulfides, selenides and tellurides, as well as arsenides and antimonides, of the transition metals crystallize with the hexagonal nickel arsenide structure shown in Figure 8. This structure is formed by a hexagonal close packing of the metalloid atoms located in the interstices, forming a simple hexagonal array. The magnetic properties of these systems have been extensively investigated in recent years and the CrSe compound was shown to have an anomalous temperature dependence of susceptibility. Hence, a neutron diffraction study of this system was undertaken.

Powder diffraction patterns were obtained at room temperature,  $77^\circ \text{K}$ , and  $4.2^\circ \text{K}$ . The room temperature pattern was in good agreement with

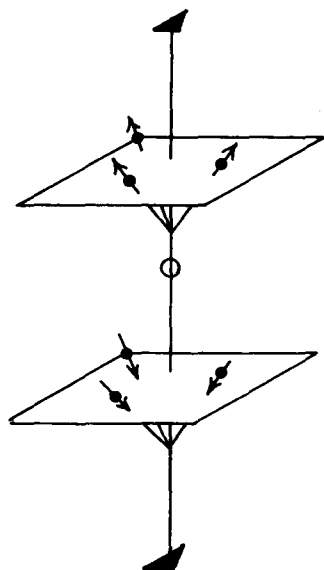


Figure 10. "Umbrella" arrangement of magnetic moments of CrSe in the  $Z=0$  and  $Z=1/2$  planes.

the calculated integrated intensities for a nickel arsenide structure. The liquid nitrogen as well as liquid helium temperature patterns showed a series of well developed superstructure lines indicating an antiferromagnetic transition. The indexing of these new peaks requires an enlargement of the chemical unit cell as shown in Figure 9.

The observed magnetic intensities can be used to rule out unambiguously all possible collinear models. This leads to the consideration of more general models in which the individual magnetic moments are no longer restricted to being parallel and antiparallel to a single direction in the crystal. A systematic treatment of the most general non-collinear model which preserves the hexagonal symmetry of the nickel arsenide structure leads to a so-called umbrella model, shown in Figure 10. The available data can give only the component of the magnetic moment in the basal plane, which turns out to be  $2.90 \mu_B$ . If a "spin-only" value for  $\text{Cr}^{+3}$  is assumed, this would correspond to an angle of inclination of about  $45^\circ$  with respect to the basal plane.

#### Single-Crystal Neutron Diffraction

The fluosilicates of divalent transition metal ions crystallize as hexahydrates, the metal ion being surrounded by a nearly regular octahedron of water molecules. Since these ions are thought to be similarly coordinated in aqueous solution, it is

of great interest to study in detail the geometrical structure and the thermal vibrational behavior of these salts. A complete structural determination of  $\text{FeSiF}_6 \cdot 6\text{H}_2\text{O}$  by single-crystal neutron diffraction has been completed as the first in a series of studies of crystalline hydrates by this technique. The fluosilicates crystallize in the rhombohedral system with a slightly distorted cesium chloride structure. The structure demands that both the  $\text{SiF}_6^-$  and  $\text{Fe}^{++}(\text{H}_2\text{O})_6$  octahedra possess threefold axes, but the octahedra may be stretched or compressed along these axes. Nevertheless, the  $\text{SiF}_6^-$  octahedron, with a Si—F distance of  $1.706 \pm 0.009 \text{ \AA}$ , is quite regular. The  $\text{Fe}^{++}(\text{H}_2\text{O})_6$  octahedron, on the other hand, is significantly stretched along the threefold axis. The two crystallographically distinct O—Fe—O angles are  $88.64^\circ$  and  $91.36^\circ$ , the difference between these angles being  $2.72^\circ \pm 0.68^\circ$ . The Fe—O bond distance is  $2.146 \pm 0.012 \text{ \AA}$ . The water molecules are oriented so that the lone pair orbital is directed approximately toward the central iron atom. Although there is some distortion from this ideal orientation with the two-fold axis directed toward Fe (the Fe—O—H angles are  $120.7^\circ \pm 1.9^\circ$  and  $126.7^\circ \pm 1.1^\circ$ ), the sum of the angles around oxygen is  $359.2^\circ \pm 1.2^\circ$ , which indicates that the system  $\text{Fe} \cdot \text{H}_2\text{O}$  is planar. The O—H bond lengths are  $0.988 \pm 0.051 \text{ \AA}$  and  $0.983 \pm 0.028 \text{ \AA}$ , and each H participates in an O—H...F hydrogen bond at a distance of  $2.702 \pm 0.014 \text{ \AA}$  or  $2.742 \pm 0.014 \text{ \AA}$  at angles of  $161^\circ \pm 10^\circ$  and  $168^\circ \pm 2^\circ$ .

The parameters describing thermal motion have also been determined. The root mean square amplitudes of vibration have been determined to an accuracy of 10%, and the directions of the principal axes of vibration to about  $10^\circ$  to  $20^\circ$ . The Si and Fe atoms vibrate isotropically. The results for O, H, and F agree with the expectation that one principal axis is always approximately parallel to a bond direction (Si—F, Fe—O, O—H). For F and O, the axes perpendicular to the bond are almost the same length and hence not well fixed in direction. For H, however, one of the other axes is perpendicular to the  $\text{H}_2\text{O}$  plane, and the third is in the plane and perpendicular to the O—H bond. The motion of H may thus be described in terms of bond stretching or in-plane bending, and an out-of-plane bending. A partial view of the structure is given in Figure 11.

In a further application of neutron diffraction, powder diffraction patterns of  $\text{HCrO}_2$  and  $\text{DCrO}_2$

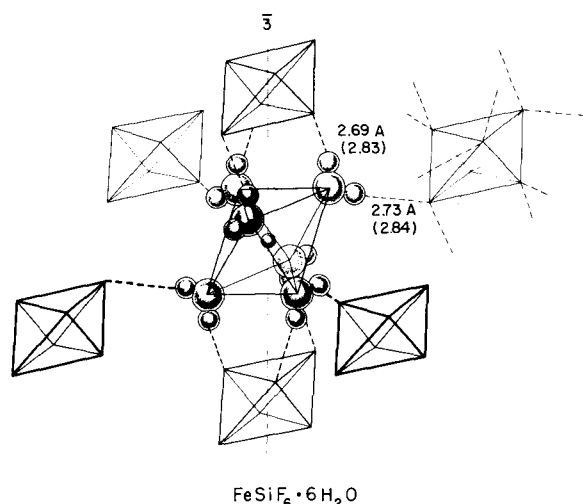


Figure 11. Partial view of the crystal structure of ferrous fluosilicate. The central  $\text{Fe}^{++}(\text{H}_2\text{O})_6$  octahedron is surrounded by eight  $\text{SiF}_6^-$  octahedra at the corners of a rhombohedron with an apex angle of  $97^\circ$ . For clarity, only six of these octahedra are shown. Each fluorine atom participates in two  $\text{O} \cdots \text{H} \cdots \text{F}$  hydrogen bonds, one linking octahedra parallel to the three-fold axis, the other linking octahedra in sheets perpendicular to the three-fold axis.

have been obtained in an attempt to determine whether the short  $\text{O} \cdots \text{O}$  distance of 2.5 Å is a symmetrical hydrogen bond. The neutron diffraction data cannot be interpreted unambiguously, but, together with nuclear magnetic resonance data from another laboratory and total neutron scattering cross sections, they seem to indicate that the H atom is disordered perpendicular to the  $\text{O} \cdots \text{O}$  bond and sits neither in a potential minimum at the midpoint of the  $\text{O} \cdots \text{O}$  line nor in two equivalent positions along this line at a more normal bond length.

## MISCELLANEOUS SUBJECTS

### Ion-Molecule Reactions

Ion-molecule reactions in the gas phase are generally characterized by much larger cross sections than those estimated from molecular structure studies. These large cross sections have been explained by the assumption of a reaction mechanism that depends on the long-range interaction of an ion and an induced dipole in a neutral molecule. In this Laboratory, the ion-molecule reactions of isotopic hydrogen giving  $\text{H}_3^+$  or isotopically substituted  $\text{H}_3^+$  have been investigated in a mass spec-

trometer. The reactions of the isotopically pure gases  $\text{H}_2$  and  $\text{H}_2^+$ ,  $\text{D}_2$  and  $\text{D}_2^+$ , and  $\text{HD}$  and  $\text{HD}^+$  are adequately treated by a model based on the above assumption. However, reactions of mixtures (such as  $\text{H}_2$  and  $\text{D}_2^+$  or  $\text{D}_2$  and  $\text{H}_2^+$ , giving  $\text{D}_2\text{H}^+$  and  $\text{H}_2\text{D}^+$  as products) show isotope effects not predicted by earlier theoretical analyses. The existence of such isotope effects may be explained in terms of zero-point energy differences in activated  $\text{H}_4^+$  complexes. They point up the importance of steps in the ion-molecule reaction mechanism which are independent of the initial long-range attraction of ions and induced dipoles in neutral molecules.

The dissociation of the hydrogen molecule-ion induced by collision with inert gas molecules has recently been investigated in other laboratories. A broad peak at  $m/e=0.5$ , corresponding to protons with half the kinetic energy of the reactant ion, has been observed in the mass spectrograph of such mixtures. There are some discrepancies in the reported cross sections for this reaction which may be caused by the neglect of ion losses in the magnetic analyzer.

For this reason, and because of a general interest in ion-molecule reactions, the analogous dissociation of  $\text{HD}^+$  induced by helium, neon, and argon was studied. A mass spectrometer with a differentially pumped ion source and an inlet system for the introduction of scattering gases was used. The integrated peak intensities at  $m/e=0.3$  and 1.3, corresponding to metastable  $\text{D}^+$  and  $\text{H}^+$  ions, were determined.

The use of  $\text{HD}$  rather than  $\text{H}_2$  facilitates the determination of the ion collection efficiency, since any discrimination in favor of  $\text{H}^+$  over  $\text{D}^+$  will appear in the relative peak intensities. It was found that there was a loss of 30% of the ion product between the scattering chamber and the collector slit.

With  $\text{HD}^+$  ions of 800-v energy,  $\text{H}^+$  and  $\text{D}^+$  ions were detected, and the yield was found to increase with increasing  $\text{HD}^+$  velocity. From a knowledge of the mass spectrometer geometry and the collection discrimination, it is possible to calculate the ratio of the transverse and forward velocities of the ion products. This ratio was found to be 0.0024 for 4250-v  $\text{HD}^+$  ions, which indicates that the transfer of energy to the scattering gas is negligible. This result also indicates that the dissociation products are not produced by a transition from the ground state of the  $\text{HD}^+$  ion to a repulsive state with the same internuclear distance. Rather, this

vertical transition must take place from a vibrationally excited state of  $\text{HD}^+$  produced by electron impact, or else only ions with a specific orientation are readily excited to dissociation.

The data on the ion losses in the spectrometer make it possible to obtain the following figures for the dissociation cross section of 4250-v  $\text{HD}^+$  ions: with He,  $1.7 \text{ \AA}^2$ ; Ne,  $2.1 \text{ \AA}^2$ ; Ar,  $2.5 \text{ \AA}^2$ . These results agree with the corresponding value for the  $\text{H}_2^+$ -He cross section.

### The Kinetics of Electron Transfer Reactions

The rate of the electron exchange reaction between thallium(I) and thallium(III) has been measured in solutions containing varying amounts of bromide with the aim of ascertaining the kinetic behavior of the various thallium bromide complexes. At constant acidity, the results can be represented by the rate law  $R = k_0(\text{Tl}^+)(\text{Tl}^{+3}) + k_2(\text{TlBr}_2^+) + k_3(\text{TlBr}_3) + k_4(\text{Tl}^+)(\text{TlBr}_4^-) + k_6(\text{TlBr}_2^-)(\text{TlBr}_4^-)$ . At  $30^\circ\text{C}$ ,  $(\text{H}^+)$  and ionic strength 0.5, the second-order rate constants  $k_0$ ,  $k_4$ , and  $k_6$  are, respectively,  $0.69$ ;  $4.6$ ; and  $2.37 \times 10^3 \text{ M}^{-1} \text{ hr}^{-1}$ . The first-order rate constants  $k_2$  and  $k_3$  are  $8 \times 10^{-3} \text{ hr}^{-1}$  and  $4.5 \times 10^{-3} \text{ hr}^{-1}$ .

The first-order terms in the rate law suggest reversible redox equilibrium mechanisms through the reactions



From the measured values of  $k_2$  and  $k_3$ , data on the potentials of the thallos-thallic and bromide-bromine couples, and the stability constants of the complexes, the values of  $k_{-2}$  and  $k_{-3}$  are calculated to be  $2.1 \times 10^7 \text{ M}^{-1} \text{ hr}^{-1}$  and  $1.2 \times 10^{11} \text{ M}^{-2} \text{ hr}^{-1}$ , at  $25^\circ\text{C}$  and ionic strength = 3. The rate of oxidation of thallos ion by bromine was measured directly by a rapid mixing and flow technique combined with spectrophotometric analysis. The values of  $k_{-2}$  and  $k_{-3}$  found are  $0.7 \times 10^7$  and  $1.4 \times 10^{11}$ , in excellent agreement with the interpretation of the exchange data.

The rapid flow-spectrophotometric technique has also been used to study the kinetics of the oxidation of the iron(II) ion by a number of substituted phenanthroline, dipyridine and tripyridine complexes of the iron(III), ruthenium(III), and osmium(III) ions. It has been found that a linear relation exists between the free energies of activation and the standard free energy changes of the

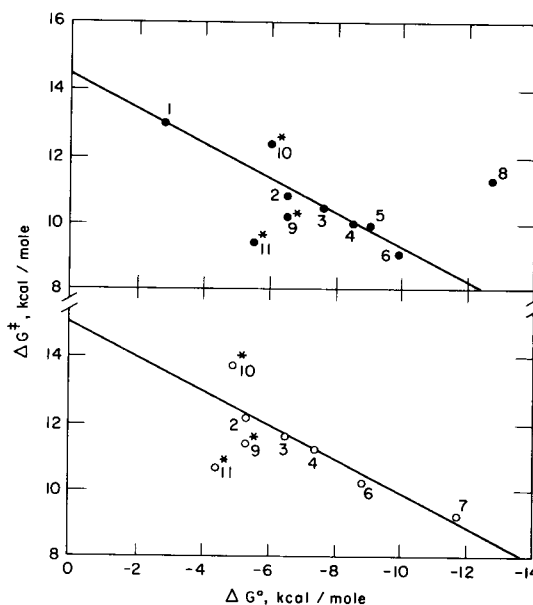


Figure 12. Relation between the free energy of activation and the standard free energy change of the reaction. o, in  $0.5 F \text{ HClO}_4$ ; ●, in  $0.5 F \text{ H}_2\text{SO}_4$ . An asterisk indicates systems possessing structures differing from the phenanthrolines.

1. *tris*-(3,4,7,8-Tetramethyl-1,10-phenanthroline)-iron(III).
2. *tris*-(5,6-Dimethyl-1,10-phenanthroline)-iron(III).
3. *tris*-(5-Methyl-1,10-phenanthroline)-iron(III).
4. *tris*-(1,10-Phenanthroline)-iron(III).
5. *tris*-(5-Phenyl-1,10-phenanthroline)-iron(III).
6. *tris*-(5-Chloro-1,10-phenanthroline)-iron(III).
7. *tris*-(5-Nitro-1,10-phenanthroline)-iron(III).
8. *tris*-(4,7-Diphenyl-1,10-phenanthroline)-iron(III).
9. *tris*-(2,2'-Bipyridine)-iron(III).
10. *tris*-(4,4'-Dimethyl-2,2'-dipyridine)-iron(III).
11. *bis*-(2,2',2''-Tripyridine)-iron(III).

reactions in which the substituted phenanthroline complexes of iron(III) are used as oxidizing agents. This is shown in Figure 12. The absence of specific steric effects in these reactions suggests that in the activated complex the ferrous ion transfers its electron to the iron(III) species once it has penetrated the space between the phenanthroline groups, rather than when it is situated on the periphery of a phenanthroline group.

### Theoretical Chemistry

The molecular virial theorem has been applied to the problem of predicting internal rotation barriers in ethanelike molecules. The data in Table 3 demonstrate that the barrier heights can be accounted for by considering only the inter-

Table 3

Rotational Barriers and Internuclear Repulsion Energies  
(in kcal/mole)

Molecule	$\Delta W^*$ (exptl.)	$\frac{1}{2} \Delta V_{nn}$ (calcd.)
$C_2H_6$	2.7-3.0	2.6
$CH_3NH_2$	1.9	2.1
$CH_3OH$	1.1	0.9
$H_3C-C\equiv C-CH_3$	0.0	0.0

\* $W$  = total molecular energy.

nuclear repulsion energy,  $\Delta V_{nn}$ . This implies that there is a cancellation of the electron-electron and electron-nucleus interaction terms. It may also be shown that a model which does not allow for a change in electronic kinetic energy would predict no rotational barrier. Electrostatic models using nonoverlapping charge distributions may serve as successful models for rotational barrier calculations, but they do not necessarily indicate the true charge distribution.

Sum rules for the transition probabilities of spontaneous emission have been derived for both atomic and molecular systems. Only electric dipole transitions are considered. In the atomic case, the sum of transition probabilities is given by the product of the nuclear charge and the electron density of the state involved in the transition. A consequence of this rule is that the ground electronic state of any atom must have a nonvanishing electron density at the nucleus. For a molecule whose vibrational potential energy can be expressed in quadratic form, the sum rule states that the sum of the vibrational transition probabilities is equal to the sum of the vibrational force constants (in suitable units).

As part of a long-range program designed to obtain a detailed theory of some simple three-atom chemical reactions, a number of programs for the evaluation of molecular integrals, in particular three-center integrals, have been written for the IBM 704 computer. These programs will be used in the *ab initio* calculation of wave functions and potential energy surfaces for such systems as  $He+H_2^+$ ,  $H_3$ , etc.

### Studies on Ancient Glass

A program of study of the composition of archaeological artifacts is being carried out by means of emission spectroscopy, flame photometry, x-ray fluorescence, and neutron activation analysis. The aims are to establish correlations between composition and provenance, dates of manufacture, methods of ancient technology, and related questions. A survey has been completed in which the concentrations of 26 elements in  $\approx 200$  specimens of ancient glass were determined. The main results and conclusions follow.

The specimens could be divided into five major compositional groups. The first group is a soda-lime glass with a high magnesium content which is now known to be found in graves of the second millenium B.C. in Egypt, Greece, Crete, Asia Minor, Mesopotamia, and Persia. The uniformity of composition of glass over this wide area reveals a high level of technological communication at this early period.

During the period of classical Greece and the great Achaemenid Persian Empire a glass of different composition was made, one of its characteristics being a relatively high antimony content. Glass of this type has not been previously reported in the literature.

With the development of the Roman Empire came a third type of glass, similar to the earlier Greek-Persian glass except that the concentration of antimony is reduced and that of manganese is correspondingly increased. This glass was made with surprising compositional consistency throughout the Roman Empire, from Egypt to the Rhineland. To the east of the Empire, however, glass of the Greek-Persian type continued to be produced. With the development of the Islamic Empire in the Near East, yet another type of glass was made, which had the high potassium and magnesium concentrations characteristic of the first type of glass (second millenium B.C.). Some specimens of a previously unreported Islamic glass containing slightly more than one-third lead oxide were also encountered.

In some cases small regional differences have become apparent within the main glass groups which permit a closer identification of the provenance of glass objects.

# Nuclear Engineering

The research activities of the Nuclear Engineering Department are chiefly concerned with the development of new reactors, reactor systems, or system components, with the over-all objective of furthering the use of all forms of nuclear energy. During this past year the Liquid Metal Fuel Reactor (LMFR) project has been discontinued, with the result that there has been considerable change of emphasis in the work of the Department. By the middle of the year, practically all work on this U-Th-Bi system had ceased, with the two exceptions of some basic chemistry studies on fused salts and the operation of the In-Pile Radiation Loop. This loop is scheduled to run until November 1960, when it will have completed a 3000-hr in-pile run. The construction of the large Utility Test Loop was completed and, after a successful trial run by BNL, the Babcock & Wilcox Company took over operation of the loop and successfully completed 1700 hr of operation at a 1000°F top temperature, about one-half the time at a 1.2-Mw power level. This highly successful run and the satisfactory operation of the other large U-Bi loops during the first half of this year increased our confidence in the over-all feasibility of the LMFR concept.

The fact that the LMFR project accounted for more than half the Department's effort during fiscal 1959 but constituted only a small percentage of the total effort at the close of fiscal 1960 has brought about considerable reassignment of personnel. Fortunately, the Department was able to do this by transferring a relatively small number of technicians to other departments in the Laboratory, and by initiating several new programs, chiefly in the fields of high temperature, liquid metals, or both. These programs include an experimental study of the release of fission products from fuel elements at elevated temperatures in a range that includes the melting point of the cladding, and the development of equipment for examining the structure of refractory nuclear fuel materials at temperatures of 2000°C or higher, as well as of the high temperature measuring devices needed for this type of work. The Department has also started a program on the study of materials

for systems containing mercury or sodium, and eventually other metals, up through the boiling point, and also will extend liquid metal heat transfer studies up through these temperatures. The Department is also extending the graphite studies to consider problems involving the testing and production of graphite for use at high temperatures. Other continuing programs include experimental and theoretical reactor physics, reactor evaluation studies, reactor chemistry, radiation chemistry, radioisotope production development, radiation effects on graphite and other reactor materials, fuel processing development, waste processing and disposal, radiation source development, and development of radiochemical analytical techniques. The service operations of the Department, including gamma irradiation, special radioisotope production, and radiochemical and metallographic analyses, and the Hot Laboratory operations, including waste handling and concentration, are adequately covered in other sections of the report from this Department.

The design of the Brookhaven High Flux Beam Research Reactor (HFBR) has engaged a large group from the Reactor Physics and Engineering Divisions. In addition, the Experimental Reactor Physics Group has made over 600 critical assemblies to complement this design work. The Lummus Company has been engaged as architect-engineer for Title I and Title II designs, with the assistance in the nuclear field first of the Curtiss-Wright Corp. and now of the Nuclear Division of Combustion Engineering, Inc. Title I design is expected to be ready in September 1960.

Phase I of the Nuclear Engineering Building is  $\approx 60\%$  complete, and occupancy is scheduled for the spring of 1961. Funds have been assigned to the Brookhaven Area Office of the Atomic Energy Commission for the construction of the high level radiation development laboratory which has been designed and is scheduled for completion in the fall of 1961. In addition, funds have been appropriated for and design started on a small volatility studies laboratory, on an addition to the critical assembly area, which will allow greater flexibility in this work, and on a much-needed addition to

the present Metallurgy Building, which will relieve the present overcrowded conditions there.

In addition to the aid supplied to the AEC in Germantown, Md., through the Department's Reactor Evaluation Group, the Department has also undertaken a program of aid to the New York Operations Office in the Pressurized Water Reactor field. On March 1, Dr. Chad Raseman was assigned to work with the AEC Civilian Power Reactors Evaluation and Planning Branch at Germantown for a period of  $\approx 18$  months.

## **REACTOR PHYSICS, DESIGN, AND EVALUATION**

### **Reactor Physics Theory**

The work of the Theoretical Reactor Physics Group during the past year was divided as usual between basic and applied problems. The latter included studies in direct support of the Department's projects, evaluation work for the AEC, and preliminary studies of breeders and other advanced reactor concepts. The basic problems included studies of neutron thermalization, resonance absorption of neutrons, fast fission, nonlinear reactor kinetics, and similar studies.

After 30 months of operation with its final core configuration, the conversion of the BNL Graphite Research Reactor from natural uranium to enriched fuel may be considered complete. This long and difficult project, which has resulted in a unique research reactor, was carried out as a collaborative effort among members of Reactor Operations, the Metallurgy Division, and the Theoretical Reactor Physics Group. Theoretical problems arising during the past year have mainly involved the use of the reactor as a research facility.

Theoretical studies have continued in direct support of the engineering and experimental work on the HFBR. The attractiveness of this small, undermoderated,  $D_2O$ -cooled reactor for research purposes was first demonstrated by theoretical studies in 1956. Its design has been shaped by users' requirements, critical experiments, theoretical calculations, and economics, as well as by engineering considerations.

Assistance in the evaluation of reactor proposals and projects for the AEC continued during the past year. Included were the evaluation of the *MS Savannah* project, Euratom proposals, the spectral shift reactor concept, and consultation on safety aspects of the Consolidated Edison Thorium Reactor.

With the cancellation of the Liquid Metal Fuel Reactor Experiment (LMFRE), the theoretical work on this reactor has been curtailed. A subcontract with Rensselaer Polytechnic Institute involving a detailed transfer function analysis of the reactor was completed during the year. This study demonstrated the inherent stability of the LMFRE. However, in view of the vital importance of breeder reactors to the future power needs of the United States, more general studies of thermal breeders were initiated. These studies indicate that breeder reactors on the  $Th-U^{233}$  cycle are feasible with a variety of weakly absorbing moderators and with solid as well as fluid fuels. The importance of fast fission and the  $(n,2n)$  reaction in beryllium in improving the possible breeding ratio of reactors was shown.

Theoretical studies directed towards a complete theory for the resonance absorption of neutrons in reactors have been continued. With improvements in experimental measurements of resonance parameters and the improvement of analytical techniques which include Monte Carlo calculations, quantitative evaluation of resonance absorption of neutrons in important nuclides such as  $U^{238}$  has been achieved. A Monte Carlo code for the calculation of high energy events in heterogeneous reactors was also completed during the year and has been used in calculations of the fast effect in uranium and beryllium systems. The results for uranium are in general agreement with integral data obtained at Brookhaven and Bettis, although the basic cross section data are still somewhat uncertain.

Measurements of the fine structure of the thermal neutron flux in a lattice cell have been perfected over a period of years at various laboratories, including Brookhaven. The theoretical analysis of these experiments has been difficult because of uncertainties in chemical binding effects. Rigorous calculation methods are being developed, however. It has been found that the free gas model gives adequate agreement with experiment for wide graphite and  $D_2O$  lattices, but inadequate agreement for water lattices. The use of better models for neutron thermalization has been found to reduce the discrepancy in the latter case. Further basic studies of chemical binding effects have also been carried out. The importance of the distribution of neutron sources in space and energy in determining the asymptotic flux distribution of an absorbing medium has been pointed out.



The stability of reactors operating at high power and flux levels has been a subject of increasing concern. A basic study of reactor stability against xenon poisoning has been completed, and the extension of the work to include temperature effects is under way.

#### Cross Section Evaluation Center

At the request of the AEC, a small group has been established to maintain a continuous review of cross section data for reactor physicists, who will be provided on request with cross section values representing the best available adjustment between theory and measurements.

The initial staffing has given this group a strong nucleus. The first projects undertaken were reviews of data on B, B<sup>10</sup>, U<sup>233</sup>, and Dy<sup>164</sup>. The group has also been reviewing and bringing up to date the list of cross section requests which the AEC has maintained. It has been assembling files of data and cross section codes. It has been responsible for the measurement of the Dy<sup>164</sup> cross section mentioned above.

#### Experimental Reactor Physics

**Water Lattices With Metal Rods.** As reported last year, the systematic study of uranium metal rod lattices is complete. It was nevertheless thought useful to devote some time to reducing the random and systematic errors associated with some of the more vital intensive parameters. Therefore miniature lattice studies of fast fission factor and resonance escape probability were repeated; these resulted in a great improvement.

A new and better technique was developed for measuring  $\rho_{28}$ , the ratio of U<sup>238</sup> captures above and below the cadmium cutoff.

**Plate Lattices.** Exponential measurements have been started on lattices of 0.122-in.-thick plates of 1.3% enriched, unclad uranium in water.

An extensive set of studies was performed to determine the effect of the geometry of the loading on the buckling deduced from flux traverses.

**Water Lattices With UO<sub>2</sub> Rods.** Exponential experiments of the same general kind have so far been performed on four volumn ratios of stainless steel clad UO<sub>2</sub> rods in light water. The fuel density is 9.29 g/cm<sup>3</sup>, the fuel diameter is 0.500 in., and the cladding thickness is 0.028 in. The fuel is 3.0% U<sup>235</sup>. The work has included a careful study of the higher order effects which might perturb the experimental results.

**Fast Fission Factor of Isolated Rods.** A series of measurements of the fast fission factor in isolated rods has been taking place on a low priority basis over the past five years. These have now been completed and analyzed. Nine rod diameters were involved, ranging from 0.250 to 3.363 in.

The values of  $\delta_{28}$  and  $\epsilon$  have been measured.

**Homogeneous Critical Assemblies.** It was mentioned in last year's report that attempts to measure bucklings of bare homogeneous or semihomogeneous assemblies had not been very successful, presumably because of room-reflected neutrons. It is possible that previous measurements at other laboratories suffered from the same problems.

An even more basic question during the early studies arose from the assumption that asymptotic reactor theory was valid in such homogeneous systems. Any departure from asymptotic theory could introduce errors of unknown magnitude.

To investigate these questions, a thin-walled, silo-type shed has been temporarily erected in back of the critical assembly area. An aluminum framework within the shed allows critical assemblies to be constructed  $\approx 20$  ft from the floor. The reflection of neutrons from the ground and building walls will be quite small; hence it should be possible to determine accurate values for both the critical mass and the buckling. Flux separability can also be readily studied, with meaningful results. The systems to be investigated will be essentially homogeneous structures of U<sup>235</sup> with C, Be, and BeO, respectively.

During fiscal 1960 the design and construction of the silo were completed; the aluminum platform was installed; all cables were run in conduit to the critical assembly area; and most of the console was installed. Design of the critical assembly table system was begun. The first runs with graphite moderator should begin late in the fall of 1960.

**Neutron Diffusion in Graphite.** Additional pulsed neutron experiments and static diffusion experiments were carried out during the year. Combining the results of both kinds of measurements has led to conclusions of greater accuracy and worth than those drawn from experiments of either type alone, because the two methods complement each other. The static experiments provide results that in principle apply to an infinite block of moderator. They then provide a point obtainable from the time-dependent results only by rather uncertain extrapolation. On the other

hand, the time-dependent study furnishes information of a kind not derivable from the static experiments. The result of the combination is in fact a joining of the best features of each.

The final results for AA graphite corrected to a specific gravity of 1.60 were

$$\begin{aligned}\lambda_0 &= 71.2 \pm 0.9 \text{ sec}^{-1} \text{ (infinite medium decay constant),} \\ D_0 &= 2.06 \pm 0.02 \times 10^5 \text{ cm}^2/\text{sec}, \\ C &= 12.4 \pm 2.2 \times 10^5 \text{ cm}^4/\text{sec}, \text{ and} \\ \sigma_a &= 3.95 \pm 0.05 \text{ mb/atom (2200 m/sec).}\end{aligned}$$

The anisotropy discovered during the static experiments has a magnitude indicated by  $D_{||} = 1.038 D_{\perp}$ , the subscripts indicating direction relative to the graphite extrusion axis.

**Neutron Slowing-Down Times.** A small-scale attack has been made on the problems of neutron slowing-down times. The time-dependent Boltzmann equation with constant scattering cross section has been solved in a rapidly converging series solution, and the results have been applied to calculating slowing-down times for different media. A set of experiments has led to evaluation of the slowing-down time in graphite. Neutron pulses from the  $\text{Be}(\text{D},n)$  target of the 1-Mev Van de Graaff accelerator were fed into a stack of interspersed layers of cadmium and graphite, and the capture gamma-rays from the cadmium were recorded by a plastic scintillator and multichannel time analyzer. The mean slowing-down time was then calculated from the distribution of counts over the time channels. The results were

Calculated  $\bar{t} = 33.3 \mu\text{sec}$  (fundamental mode alone);  
Observed  $\bar{t} = 33 \pm 3 \mu\text{sec}$ .

**Delayed Neutron Shielding.** At the request of the Babcock & Wilcox Company, a set of experiments was undertaken to show how delayed neutrons from fissions are attenuated in four different kinds of concrete: ordinary, barytes, ilmenite, and ferrophosphorous. The results will be useful in designing shields for the external parts of primary loops of circulating fuel reactors.

The delayed neutrons were generated by exposing small amounts of  $\text{U}^{235}$  in the fast pneumatic tube of the Graphite Research Reactor. The samples were then rapidly withdrawn by pneumatic action to the center of stacked concrete shielding. Thermal neutron intensities were measured near the source by activation of indium; far from the source a  $\text{BF}_3$  counter was used.

As expected, the delayed neutrons are attenuated more rapidly than prompt fission neutrons. The latter have a higher median energy.

**Transfer Functions.** A new way of measuring reactor transfer functions has been developed. The reactivity is varied in a random way through rapid motion of a small neutron poison. The response to this variation is measured and recorded. The input and output autocorrelation functions and the cross-correlation function are calculated by passing the signals through an analogue computer. Fourier decomposition of the autocorrelation and cross-correlation functions then leads to both the amplitude and the phase of the transfer function.

The method shows promise because it can be applied safely to operating high power reactors and can lead to insight into negative feedback mechanisms and sources of instability. It will allow measurement of the low frequency end of the transfer function without the hazardous aspects of the usual oscillator techniques.

**Pressurized Critical Experiments.** According to present plans, the assembly cell in the forthcoming addition to the critical assembly facility will be used for a set of high pressure, high temperature experiments. Preliminary design of the system has begun. Procurement of components will begin late in 1960.

**Cross Section Measurements.** Cross section measurements have been undertaken in certain cases where the data desired have primary importance in reactor physics, either for reactor design or for interpretation of reactor physics experiments. The measurements made so far deal with the transmission and activation cross sections of  $\text{Dy}^{164}$ , the transmission cross section of  $\text{Eu}^{151}$ , and the activation resonance integrals of  $\text{Dy}^{164}$  and  $\text{Cu}^{63}$ . The  $\text{Dy}^{164}$  activation is that which leads to the 2.32-hr decay.

The measured transmission cross section of  $\text{Eu}^{151}$  was analyzed to yield the Breit-Wigner parameters at 0.321 and 0.460 ev.

**Reactivity Meter.** The large quantity of critical experiments (over 700) done in connection with the design of the new research reactor was only possible because of the development of a new and powerful tool for measuring excess reactivity. The system used is based on coding the BNL general-purpose analogue computer to accept a flux signal from a critical assembly and to solve instantaneously the pile kinetic equations. The quantity

solved for is the reactivity of the system. The output is taken from a curve plotter or a digital voltmeter. There is no need to wait for asymptotic periods to be achieved, or to analyze any data after the experiment is finished. The results, accurate to about  $\delta\rho = 10^{-5}$ , are generated as the experiment is being performed.

The reactivity meter has been used in measurements of danger coefficients as a function of position throughout the core and the reflector of the HFBR critical assemblies. It has also been used in complete calibrations of control rods with total worths of as much as 12%  $k$ . The time required for such a calibration depends only on the rod speed, since the calibration curve appears on the curve plotter as the rod or rods are driven in. This system has now been used steadily and satisfactorily for  $\approx 1$  yr.

**Advanced Research Reactors.** A small-scale program on several promising new single-purpose research reactor concepts has begun. Primary attention has been given so far to different versions of pulsed reactors. Preliminary design of an  $\text{H}_2\text{O}$ - $\text{U}^{235}$  solution system has been started. Survey calculations are being run on an alternative design which would contain a solid homogeneous mixture of  $\text{U}^{235}$  and  $\text{MgO}$ . The solution reactor could be run as a circulating fuel system with external heat removal and frequent pulsing. The solid system seems to possess the advantage of being able to achieve pulses of very high integrated power.

Either kind of reactor would be extremely useful in the study of relaxation phenomena of all kinds, physical, chemical, and biological.

**HFBR Critical Experiments.** During the year  $\approx 700$  critical experiments have been performed on various versions of the HFBR. In most of these the core or structural components or both had been changed from those of a previous state, and the experiment was done to determine how the reactivity was affected. Other experiments gave information on problems not primarily related to reactivity (power and flux distributions, neutron lifetimes, etc.).

The goal of this experimental program differed in a very basic way from that of most others undertaken at BNL. Concern here was not with helping to improve the status of reactor theory, but with arriving at recommended properties of a research machine that is passing through the design stage. The experimental program was therefore oriented toward experimental optimization.

The theory was used as a guide to the experiments, instead of the converse.

These measurements have assisted in arriving at a design that (1) has very nearly the right excess  $k$  to support the planned operating cycle, (2) has the least reactivity-depressing structure in the core vessel, (3) has a beam tube layout optimized for intensity and background, and (4) will more than meet advance estimates of output neutron flux. The sources of reactivity transient hazards have been studied carefully. The heat source aspect of the heat removal problem is on a sound experimental footing. The safety rod properties are known.

A rough mock-up containing the core and structural components now incorporated in the design has been assembled and tested. A final and precise mock-up is being procured. When the exact mock-up has passed through a complete set of measurements of excess reactivity, power distribution, flux distribution, poison reactivity effects, etc., the neutron performance to be expected from the operating HFBR will be accurately known.

Some of the fundamental results of the measurements to date are

Design core volume	91 liters
Total rod worth	36% $k$
Standard holddown by rods at start of a fuel cycle	26% $k$
Neutron lifetime	$672 \pm 13$ $\mu\text{sec}$
Peak-to-average power in the cold clean core	$3.50 \pm 0.2$
Peak thermal neutron flux	$1.9 \times 10^{13}$ n/cm <sup>2</sup> -sec/Mw

The expected peak neutron fluxes given in the last annual report were based on preliminary calculations of the reactor properties. These calculations left no allowance for structural effects or for the strong power peaking that has been observed, which have the effect of reducing the calculated fluxes. With the introduction of more realistic design considerations it is now clear that HFBR fluxes will exceed those in the original proposal by only  $\approx 50\%$ .

The critical experiments have required the development of a number of new techniques. The most important of these stemmed from the need to measure the excess reactivity of highly supercritical assemblies. The procedure adopted involved poisoning the assembly core to the just supercritical state with a uniform distribution of neutron absorber. The uniform differential poison worth was measured, and the total poison worth was then found by recourse to a simple mathematical

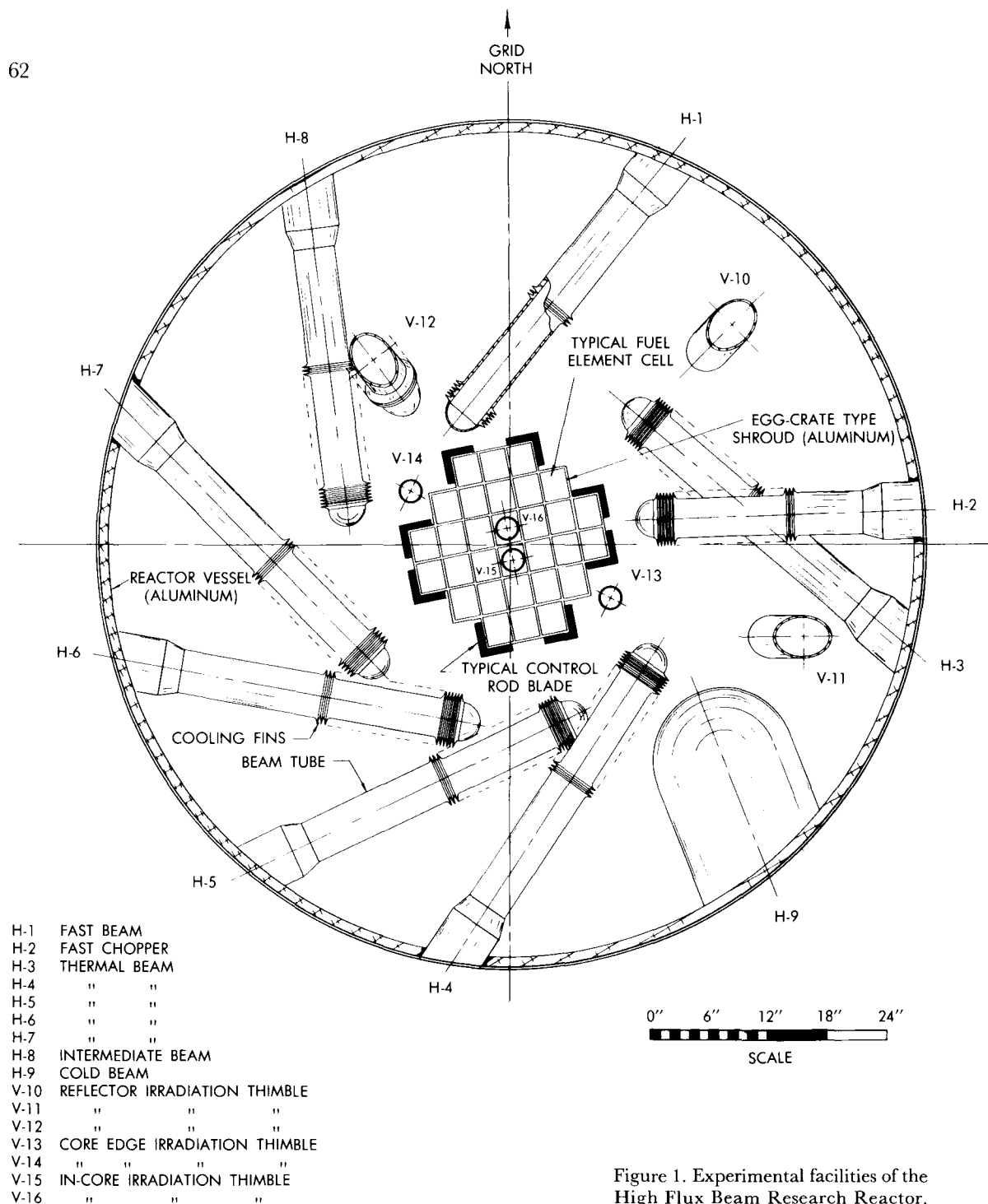


Figure 1. Experimental facilities of the High Flux Beam Research Reactor.

model which relates the differential worth to the integral worth in this reactor.

#### Brookhaven High Flux Beam Research Reactor (HFBR)

During the past year all the essential features and most of the minor components of the design for the HFBR have been fairly well determined by the staff of the reactor project aided by the

critical assembly studies and the architectural engineering group. Title I design should be completed by the end of August.

A brief description of the principal features of the reactor and reactor building according to the present design follows.

The reactor is cooled, moderated, and reflected with heavy water. Figure 1 shows the experimental facilities of the HFBR, and Figure 2 is a section

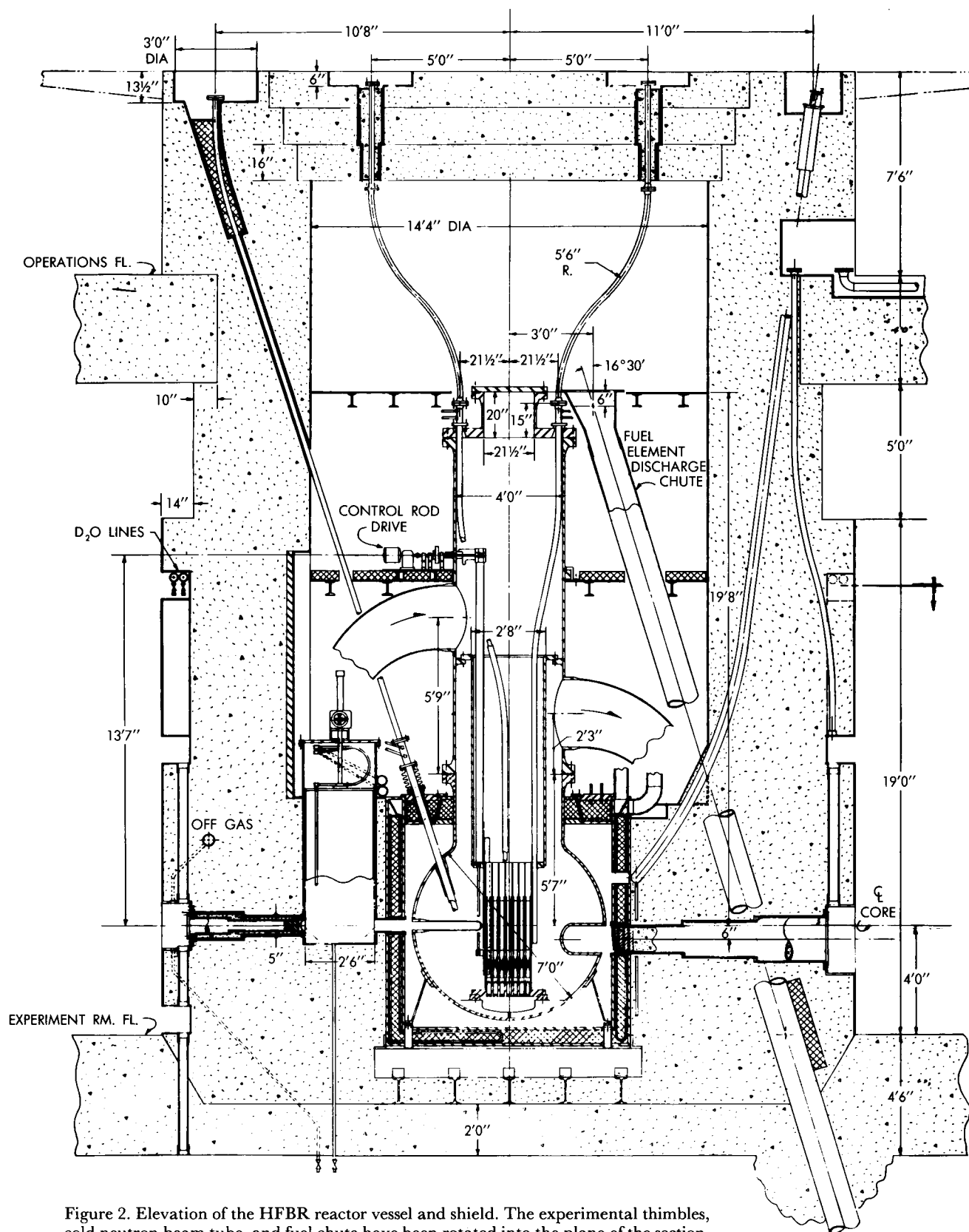


Figure 2. Elevation of the HFBR reactor vessel and shield. The experimental thimbles, cold neutron beam tube, and fuel chute have been rotated into the plane of the section.

through the core, core vessel, thermal shield, and biological shield, showing the arrangement of the principal parts.

The core proper, composed of 28 fuel elements of the type used in the Engineering Test Reactor, is located in a shroud at the center of the lower

spherical part of the reactor vessel. Cool water from the D<sub>2</sub>O pumps is delivered into the upper cylindrical part of the reactor vessel, flows downward inside the shroud, through the fuel elements, and is discharged into the bottom of the spherical vessel. It then flows upward in the spherical vessel

Table 1

## HFBR Design Parameter List

Power	40 Mw	Core thermal design data	
Neutron flux, n/cm <sup>2</sup> -sec		Water channel thickness, in.	0.105
Core, total epithermal	1.6×10 <sup>15</sup>	Fuel plate thickness, in.	0.050
Reflector thermal flux, max	7×10 <sup>14</sup>	Fuel plate length, active, in.	21
Materials		Water channel width, in.	2.231
Coolant, moderator, and reflector	D <sub>2</sub> O	Core alloy width, in.	2.00
Fuel	U <sup>235</sup> -Al alloy	Heat transfer surface per element, ft <sup>2</sup>	11.1
Core structure, beam tubes	6061 Al	Total heat transfer surface, core, ft <sup>2</sup>	311
Lower vessel	6061 Al	Water flow data	
Upper vessel	Stainless steel	Water channel flow area per element, ft <sup>2</sup>	0.0326
Primary pipes & process equipment	Stainless steel	Total channel flow area, core, ft <sup>2</sup>	0.912
Fuel element and core		Water velocity in channels, ft/sec	35
Type	ETR, flat-plate	Water flow per element, gal/min	520
Uranium concentration in core alloy, wt %	30	Water flow, 28 elements, gal/min	14,560
Core thickness, in. U-Al	0.020	Water flow in control rods, gal/min	≈2,000
Cladding thickness, in. X 8001 Al	0.015	Water flow in bypass, gal/min	≈1,000
Fuel region dimensions per element, in.	2.6×3.0×21	Total water flow, gal/min	≈17,600
U <sup>235</sup> loading per element, gal/min	260	Total primary loop pressure drop, psi	60
Number of fuel elements	28	Pressure drop in fuel elements, psi	25
Core volume, liters	86	Process system design	
Total U <sup>235</sup> loading, kg	7.28	Maximum operating pressure, psig	250
Water-to-metal volume ratio	1.23	Maximum operating temperature, °F	150
D/U <sup>235</sup> atom ratio	169	Vessel design temperature, °F	200
Al/U <sup>235</sup> atom ratio	123	Beam tube design temperature, °F	400
Cycle time for 20% burn-up, days	40	Core power conditions	
Temperature and void coefficients		Radial peak-to-average power density	1.73
Core metal, % k/°C	-0.36×10 <sup>-3</sup>	Over-all peak-to-average power density	3.0
Core water, % k/°C	-6.15×10 <sup>-3</sup>	Bulk hot channel factor	1.36
Reflector water, % k/°C	-10.5×10 <sup>-3</sup>	Film hot channel factor	1.87
Total temperature, % k/°C	-17.01×10 <sup>-3</sup>	Average power density, Mw/l	0.465
Core void, % k/cm <sup>3</sup>	-0.33×10 <sup>-3</sup>	Maximum power density, Mw/l	1.40
Excess reactivity requirements, %		Core thermal analysis results	
Burn-up, plus Sm and stable FP's	4.3	Core inlet temperature, °F	120
Xenon, steady state	4.0	Outlet temperature, °F	133.4
Temperature	0.5	ΔT bulk, °F	13.4
Control and experiments	1.2	Hot spot surface temperature, °F	356
Total	10.0	Hot spot saturation pressure, psig	131
Control rods		Hot spot pressure, actual, psig	173
Number of main rods	8	Thermal margin from hot spot to saturation, °F	20
Individual main rod, total worth, % k	4.8	Blanket gas pressure, psig	196
8 main rods in gang, total worth, % k	32	Average heat flux, Btu/hr-ft <sup>2</sup>	0.387×10 <sup>6</sup>
Number of auxiliary rods	8	Maximum heat flux, Btu/hr-ft <sup>2</sup>	1.16×10 <sup>6</sup>
Individual auxiliary rod, total worth, % k	1.0	Burn-out heat flux, Btu/hr-ft <sup>2</sup>	3.7×10 <sup>6</sup>
8 auxiliary rods in gang, total worth, % k	6		
Total worth of all rods, % k	38		

outside the core and leaves from the outlet pipe on the upper cylindrical part of the vessel.

The warm  $D_2O$  flows through coolers in the basement of the reactor building, where the heat added in the reactor is removed by exchange with water from cooling towers.

The fuel element design and the hydraulic and heat transfer requirements are patterned closely after existing practice at the Materials Testing Reactor. No special fuel element development program is planned.

Table 1 is a presentation of the principal design parameters for this reactor.

The spherical portion of the reactor core vessel is surrounded by a thermal shield. This is a water-cooled, tanklike structure composed of steel plates and lead cadmium alloy. The thermal shield protects the concrete biological shield, which is of conventional design, from the gamma and neutron flux leaking from the core vessel, and provides an emergency containment to keep the reactor core covered with heavy water in event of a leak in the primary vessel.

The 16 control rods provided for the reactor are located in the reflector, a few inches outside the core, where the thermal flux peak is found. All are driven by drive mechanisms located on the upper portion of the reactor vessel. Eight main rods are driven downward to blanket the reactor core, while eight auxiliary rods are pulled upward and aligned with the bottom of the core, for the "all in" position.

Normal approach to critical will occur when the eight main rods are pulled upward so that their bottom edges are about even with the top of the core. Thereafter, as the fission product poisons accumulate in the fuel, the auxiliary rods are dropped and the main rods are pulled up equal distances to maintain the flux peak constant on the beam tubes.

The fuel is changed on a 20-day cycle. In this operation the 14 peripheral elements are removed from the top of the reactor vessel and are passed into the fuel chute for delivery to the canal in the basement. The 14 center elements are then moved to the periphery of the core, and 14 new elements are inserted in the center.

The reactor is housed in a three-story circular domed building which will be reasonably gas tight. Concrete or steel construction is contemplated for this building. The bottom floor will contain the machinery needed for operation of the reactor.

The second or ground-level floor is reserved for beam experiments and laboratories. The top floor is for irradiation experiments and fuel handling, and the control room will be located here.

### Evaluation and Advanced Design Studies

A major portion of the evaluation work during the year was concerned with a comprehensive review study of the utilization of fission fragment energy for the fixation of nitrogen. For a proper appraisal of a chemonuclear system, studies in five areas were necessary, i.e., radiation chemistry, fuel element configuration, reactor design, chemical process design, and economic evaluation.

The fuel types studied were dust or circulating fuels and fixed fuels. Fixed fuels were further classified as fibers, plated porous beds, or filaments.

The main objective of the studies was assessment of the technical problems that must be solved in developing such systems. Reports covering this work will be issued during the latter part of 1960.

A survey was undertaken of the status and potential of direct conversion in this country, with emphasis on civilian nuclear power application. Four major approaches were covered: thermoelectric, thermionic, magnetohydrodynamic, and fuel cells. An unclassified version of the report on this work will be completed within the next few months.

A limited amount of theoretical work on direct conversion of fission energy to electrical energy was carried on. This is a long-range program of investigation on the characteristics of a fission plasma.

There are at present three reactor concepts at various stages of investigation within the Department. All three fall under the general classification of nonrigid fuels. They consist of (1) the  $UO_2$ -Na fuel reactor known as the Laminar Fluidized Bed Reactor; (2) U-Bi or U-Pb-Bi in fixed fuel elements; and (3) a granulated graphite and uranium fuel in fixed fuel elements operating at temperatures at which radiation is the dominant heat transfer mechanism. The Reactor Evaluation Group will study these concepts and in conjunction with the staff personnel involved will determine the most promising approach. Finally, a considerable number of evaluation studies were undertaken for the AEC during this period. Representative of this was the technical assistance given in evaluating the power reactor proposals submitted to Euratom in October 1959.



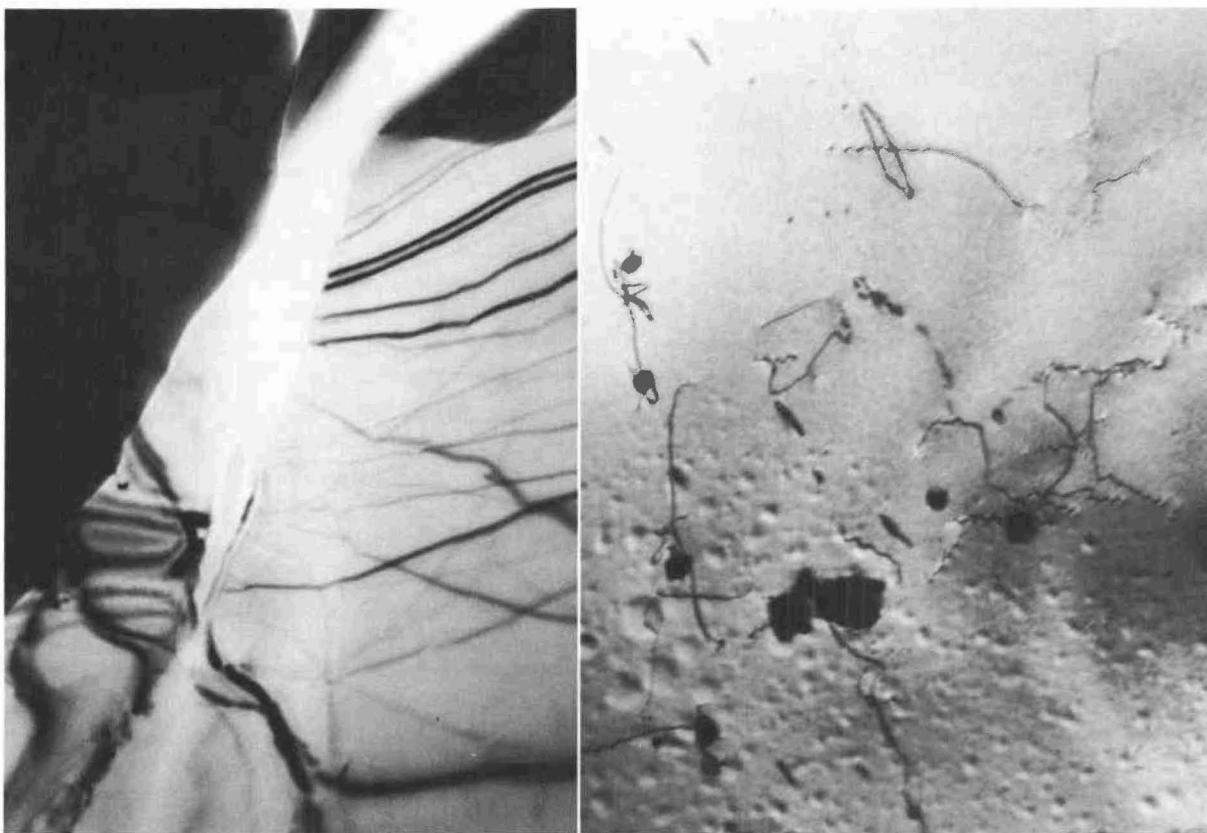


Figure 3. Electron micrographs of aluminum foil electropolished to electron transparency. Left: Unirradiated sample; magnification 50,400 $\times$ . Right: Sample irradiated in the Graphite Research Reactor for a total exposure of  $2 \times 10^{17}$  nvt; magnification, 13,680 $\times$ . Note larger number of irregular dislocations with helical configurations, loops, etc. Background structure is also characteristic of neutron-damaged metals.

## METALLURGY

### Materials Development

**Materials for Containing Pb-Bi Eutectic.** As the LMFR project was closed down, work was started on developing materials for other liquid metal systems. On one such system, the Pb-Bi eutectic, some work had been done some time ago. Several Pb-Bi eutectic loops, including one pumped electromagnetically, were put into operation to study the corrosion and mass transfer rates of low Cr steels and carbon steels by Pb-Bi eutectic (44.5% Pb, 55.5% Bi).

**Materials for Containing Boiling Liquid Metals.** Planning was commenced on a program to study the materials problems for systems containing mercury or boiling sodium. The early phase of the boiling mercury work will consist of screening capsule tests and 10 to 12 loops operating at 1000° to

1100° F boiling and 1100° to 1300° F superheat. The loops will be made of coextruded pipe (carbon steel inside, 1¼ Cr - ½ Mo steel outside), 1¼ Cr - ½ Mo steel, 2¼ Cr - 1 Mo steel, carbon steel-Inconel coextruded tubing, and Cb - 1 Zr alloy tubing. The capsule tests will screen these materials and, in addition, molybdenum, pure iron, tantalum, high speed steels, etc. The sodium work will begin with the construction of a sodium purification system.

**Bearing Materials for Liquid Metal Service.** The bearing properties of various materials immersed in liquid bismuth were tested. Results showed ceramics such as SiN, SiC, and SiN-bonded SiC to have good wear resistance. However, those refractories containing SiC were attacked by bismuth. The tests also showed that the bismuth acts as a lubricant and that prewetting the rubbing surfaces reduces the wear and coefficient of friction.

## Fundamentals of Corrosion

The following new observations were made on the formation of inhibiting deposits on steel surfaces contacted with Zr-Bi melts.

1. Uniform film formation can best be achieved by partially restricting the access of the Bi-Zr alloy to the surface. Such restriction is achieved by closely spacing two parallel polished surfaces and is believed to prevent dross usually present on the bismuth surface from contacting the prepared surface and interfering with film formation.

2. Film growth rate studies using the above technique suggest that the film that forms during the first few hours spalls and is succeeded by zirconium deposition that follows a parabolic rate of growth.

3. The corrosion of steel samples in Bi without inhibitors at 700° to 800°C leaves carbide particles protruding from the surface which react to form ZrC upon contact with Zr-Bi melt, even on a steel on which ZrN deposits normally form.

4. The content, distribution, and activity of both carbon and nitrogen in a steel simultaneously affect its resistance to attack by inhibited bismuth.

Further work with the restricted access technique is planned to throw more light on the mechanism of growth of the inhibiting deposits.

Studies were initiated to investigate any possible relationships between electrochemical (i.e., thermocouple) effects and selected corrosion of certain materials in liquid bismuth. Electrodiffusion experiments showed that iron migrates through bismuth toward an inert (tungsten) anode, while chromium migrates toward an inert cathode. The thermocouple potentials between the various steels tested by the Materials Development Group were measured while immersed in bismuth and showed a maximum positive potential for the 5 Cr steel against 2¼ Cr - 1 Mo steel. This couple has consistently produced selective attack of the 5 Cr material in loop experiments. The use of magnetic susceptibility measurements to determine the electronic structure of the dissolved species has been started, and equipment is under construction.

## Metallography

An electron-micrographic investigation was begun of the mode of nucleation and growth of pit corrosion occurring in the grain boundaries of high purity iron in aqueous media. Preliminary studies of the effect of the hydrogen ion and the chloride ion were completed, and time-lapse cine-

matography was employed in measuring the rate of pit growth. At  $pH < 6.5$ , attack occurs indiscriminately along grain boundaries. At  $pH$  6.5 to 7.0, attack occurs as well-defined circular pits on high angle grain boundaries. At  $pH$  7.0, attack is nonspecific, appearing randomly over the whole specimen, both on grains and at grain boundaries.

The electron microscope was used to investigate the effects of gamma and thermal neutron irradiation of mica and of aluminum. The aluminum was thinned to  $< 1000 \text{ \AA}$  by electropolishing, and the dislocations in the aluminum lattice were directly observed. Pile irradiations up to  $10^{16} \text{ nvt}$  produced increases in dislocation densities, as seen in Figure 3, and markedly accelerated the electrochemical reaction of aluminum to the electrolytes used in the thinning operation.

A method for the study of the effects of fission fragments and other high energy particles was developed. It involves the deposition of metal films containing small, known amounts of  $U^{235}$ , which

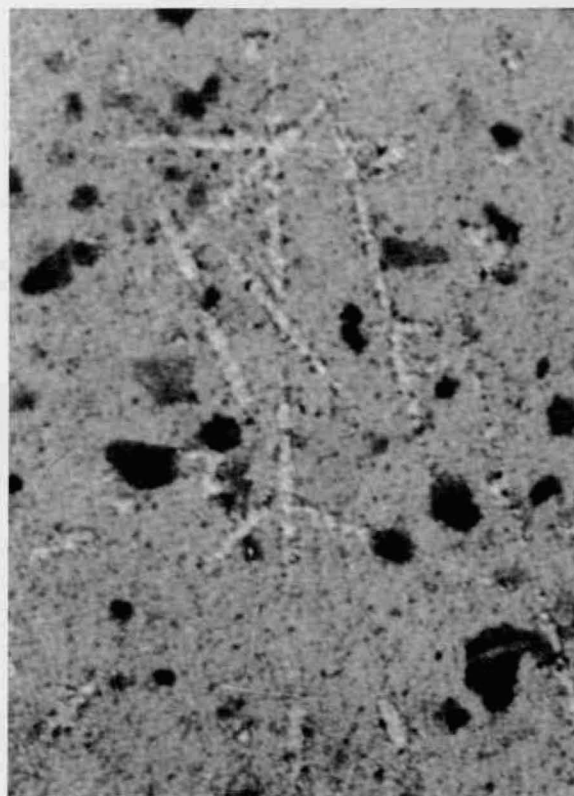


Figure 4. Electron micrograph of fission fragment tracks in evaporated film composed of an aluminum overlay on  $U^{235}$  on an aluminum substrate. Total exposure to thermal neutrons,  $2.8 \times 10^{15} \text{ nvt}$ ; magnification,  $140,000\times$ .

are irradiated in the BNL Graphite Research Reactor and then examined in the electron microscope. This technique results in direct observation of the tracks of the fission fragments, as seen in Figure 4. The mechanism of fragment damage to the matrix metal and the effects of the degree of perfection of the matrix lattice are being investigated.

Further experiments are planned to explore the physical effect of  $\alpha$ -particles, protons, and deuterons on thin metal crystals.

### Radiation Effects

A fundamental program to study the effect of neutron irradiation on the properties of iron, steel, and other body-centered-cubic metals was initiated. The objective of this program is to study the manner in which material and irradiation variables change the mechanical properties of this class of metals, in order to explain the mechanism of irradiation effects on these properties. The initial work will be on high purity iron and synthesized steels made by alloying high purity iron with the various elements generally found in steels.

Effects of irradiation on the following properties are being studied: yield strength, tensile strength, breaking strength, ductility, true stress and strain relationships, work-hardening characteristics, hardness, the ductile-brittle transition, and electrical resistivity. All these properties will be determined from unirradiated specimens, from irradiated specimens, and from specimens that have been irradiated and then annealed to study the degree of thermal recovery. Structural changes in the materials will also be studied by means of metallographic techniques, electron microscopy, and x-ray diffraction.

The Instron tensile tester has been modified to perform tests from room temperature to liquid helium temperature for remotely testing specimens from the irradiation effects program. An optical comparator has been set up to measure the diameter of the test specimens. A remotely operated electrical resistivity setup and a full-sized impact test machine were added during the fiscal year.

### Graphite Studies

The graphite program at BNL includes studies of the BNL Graphite Research Reactor radiation and anneal changes, the parameters affecting graphite oxidation and "burning," and the thermal conductivities, Brunauer-Emmett-Teller sur-

face areas, helium densities, and porosities of various graphites. In addition, a new program is being initiated to study the mechanisms of bond formation under different graphitizing conditions.

**BNL Graphite Research Reactor Studies.** The BNL annealing procedure (since May 1959) has been to maintain the maximum graphite temperatures for 4 to 8 hr, then reverse the air flow, and maintain temperature for another 4 to 8 hr. With this procedure, temperature variations throughout the reactor are minimized, and regions over 300°C extend to 6 ft from the reactor center.

Contour recovery measurements on the reactor and in-pile sample recoveries have exceeded 100% for the 10th, 11th, 12th, and 13th anneals; i.e., recovery during each anneal has been greater than growth during the period between anneals. The bidirectional air-flow method and the extended "soak times" have thus resulted in the most effective anneals achieved to date.

**Graphite Oxidation.** The results obtained from the oxidation program indicate that the oxidation rates of the AGOT graphite originally used in the BNL Graphite Research Reactor vary from sample to sample. In any one 4×4×24-in. block the rates vary by a factor of 15 and do not show any simple dependence on impurities or original density. At 400°C the oxidation rates in O<sub>2</sub> range from 6.0 to 75×10<sup>-10</sup> g/cm<sup>2</sup>-sec and are first order with O<sub>2</sub> pressure from 1/4 to 2 atm.

Ultrasonic cleaning and/or soaking in methanol often reduces the oxidation rates of small samples (1/4-in. diameter) and accelerates the rates of large samples (3-in. diameter). Ultrasonic cleaning and/or soaking in distilled water appears to increase the oxidation rate of all samples.

**Graphite Burning Studies.** Studies were made under isothermal and adiabatic conditions to determine the conditions leading to runaway in an air-cooled, graphite moderated reactor. From these studies it was possible to express empirically the length of channel required to preheat the inlet air to a temperature at which the heat generated by oxidation just equals the heat removed by the air stream. (Beyond this point the temperature will rise because of oxidation heat.)

The empirical equation best fitting the turbulent flow data is

$$X_{\text{turb}} = 1.4f/D^2 - \frac{T}{8} + \frac{5 \times 10^{-5}}{R_{705^\circ\text{C}}} + 85,$$

and the equation best fitting the laminar flow data is

$$X_{\text{lam}} = 1.4f/D^2 - \frac{T}{8} + \frac{5 \times 10^{-5}}{R_{705^\circ\text{C}}} + 86;$$

where  $X$  (ft) is the length of channel necessary to reach equilibrium between the heat generated and the heat removed,  $D$  (in.) is the diameter of the channel,  $T$  ( $^\circ\text{C}$ ) is the channel temperature,  $f$  (SCFM) is the air flow rate, and  $R_{705^\circ\text{C}}$  ( $\text{g}/\text{cm}^2\text{-sec}$ ) is the graphite reactivity, determined experimentally from oxidation rate measurements.

The results have shown that, even under unfavorable conditions, the temperature range in which graphite reactions become troublesome is  $>600^\circ\text{C}$ , which is well above the operating temperatures for the type of fuel elements used in air-cooled reactors. No unstable situation was found at temperatures  $<750^\circ\text{C}$  when the oxidation rate of the graphite (in  $\text{cal}/\text{cm}^2\text{-sec}$ ) was  $<50$  times the value of the heat transfer coefficient (in  $\text{cal}/\text{cm}^2\text{-sec-}^\circ\text{C}$ ).

#### Spatially-Fixed, Nonrigid Fuel Element

A program has been started to investigate a reactor fuel element offering possible advantages in fabrication, reprocessing, and resistance to radiation damage over other fuel elements now in use or under study. As now conceived, the element would consist of a partly or completely molten metal alloy sealed in an impervious graphite container. Alloys would be composed of fissile metals, fertile metals, and low cross section, low melting metals. Numerous possibilities exist in the U-Pu-Th-Bi-Pb-Sn alloy system. Experiments are in progress to determine the effects of anticipated thermal and radiation conditions of reactor operation on such alloys. Preliminary results indicate that imposition of a temperature differential on stagnant, partially molten U-Bi and Th-Bi alloys causes migration of uranium and thorium from hotter regions to cooler regions, probably because of the effect of temperature upon the solubility of uranium and thorium in bismuth. It has not yet been established whether this migration is due to diffusion or to eddy currents in the liquid alloy. Techniques are being developed for studying the behavior of inert fission product gases generated by irradiations in these alloys.

#### HFBR Fuel Elements

The Metallurgy Division is frequently called upon for assistance in other projects throughout the Laboratory. One of the largest such efforts this

year was concerned with the selection of materials for the HFBR.

A radiation test experiment for the HFBR fuel element material has been designed and samples have been fabricated and shipped to the Engineering Test Reactor (ETR) for irradiation. Post-irradiation examination will be performed at the Materials Testing Reactor (MTR) under the supervision of BNL personnel. This U-Al element contains a 30 wt % U-Al alloy core with 3 wt % Si added, and is clad with X-8001 Al. Burn-up will be 40 to 50% of the total uranium content. Two additional ETR test samples are being fabricated, one containing 35 and the other 40 wt % uranium in the core.

Thermal cycling tests on several fuel element designs have been performed and have proved the dimensional stability of the selected configurations.

### CHEMISTRY AND CHEMICAL TECHNOLOGY

#### Reactor Chemistry

**Thermodynamics of Alloys.** Galvanic cells have been further exploited as sources of thermodynamic data, both for solutions of metals in bismuth and for a solid,  $\text{ThC}_2$ . A variety of different cells involving this compound have been constructed in

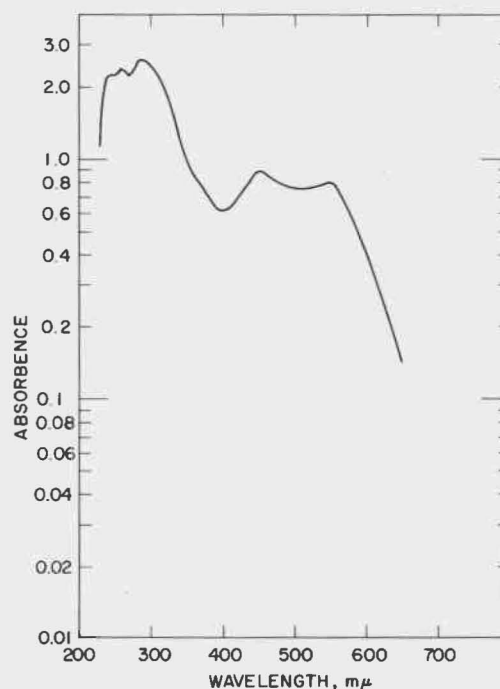


Figure 5. Absorption spectrum of  $\text{UCl}_3$  in ternary eutectic at a concentration of 1315 ppm and a slit width of 0.04 mm.



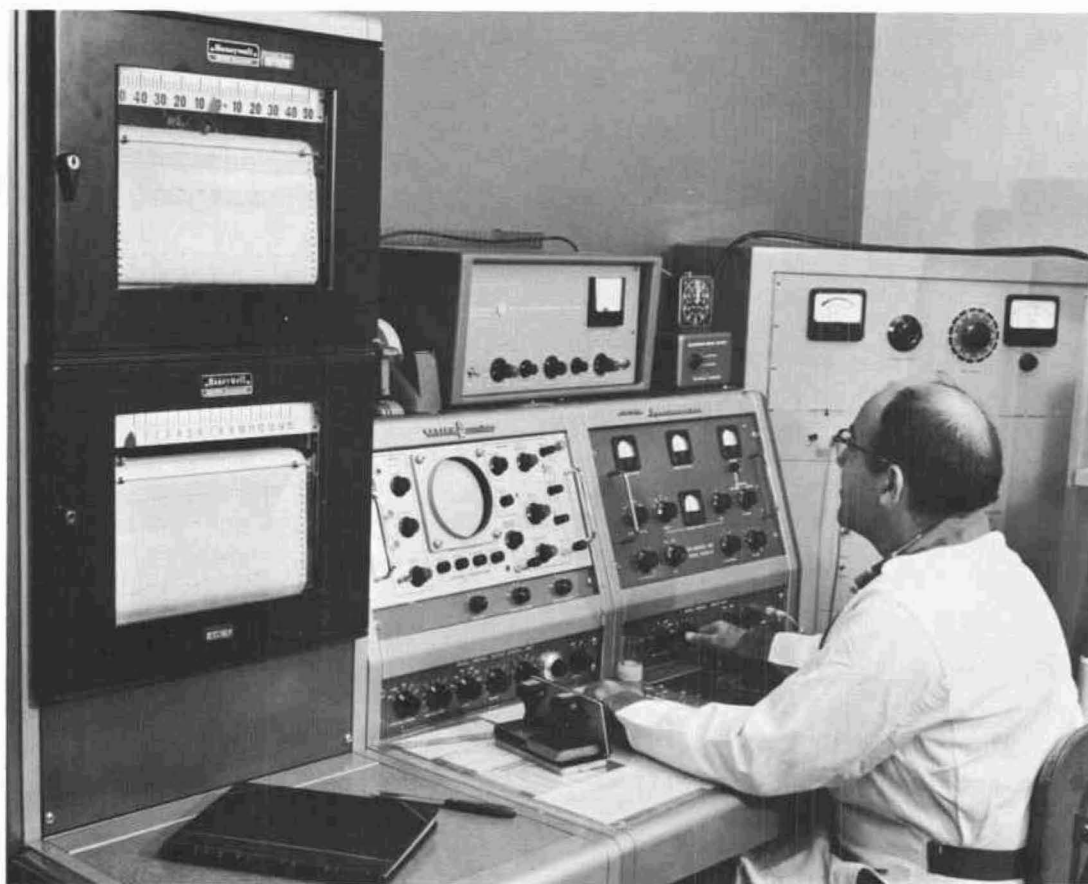


Figure 6. Operating the controls of an electron spin resonance spectrometer.

order to find a highly reliable and reproducible system, since this might open the way to quick determination of the free energies of formation of many other carbides. Fair success has been achieved with such cells as

$\text{Th} | \text{ThF}_4(5\%), \text{CaF}_2(95\%) | \text{ThC}_2, \text{C}$ ,  
and

$\text{Th} | \text{ThCl}_4(5\%), \text{LiCl} + \text{KCl}(95\%) | \text{ThC}_2, \text{C}$ .

In the former the electrolyte is solid at the experimental temperature; in the latter, liquid.

**Spectroscopy of Fused Salts.** In connection with a study of the oxidation state of uranium in fused chloride mixtures, the absorption spectrum of solutions of  $\text{UCl}_3$  in the ternary eutectic  $\text{MgCl}_2$ - $\text{NaCl}$ - $\text{KCl}$  at  $440^\circ\text{C}$  was obtained. The first solutions proved very unstable, and it was necessary to resort to rigorous purification procedures; the salt must be completely oxygen-free. A representative spectrum is given in Figure 5.

## Radiation Chemistry Research

**Solid State Polymerization.** The polymerization reaction of crystalline acrylamide has been intensively studied by using electron spin resonance (see Figure 6), x-ray diffraction, microscopy, and time-lapse photography as well as conventional solution techniques. The reaction involves several different types of free radicals with long lifetimes and is greatly affected by impurities and crystalline defects.

**Graft Copolymerization.** Mylar-styrene copolymers were prepared by various techniques, and their properties were studied. The molecular weight of the graft has been controlled by varying the solvent composition, monomer concentration, irradiation dose, irradiation temperature, and reaction temperature.

**Ionic Polymerization.** Attempts are being made to clarify the phenomena of ionic polymerization initiated by irradiation. These feature a

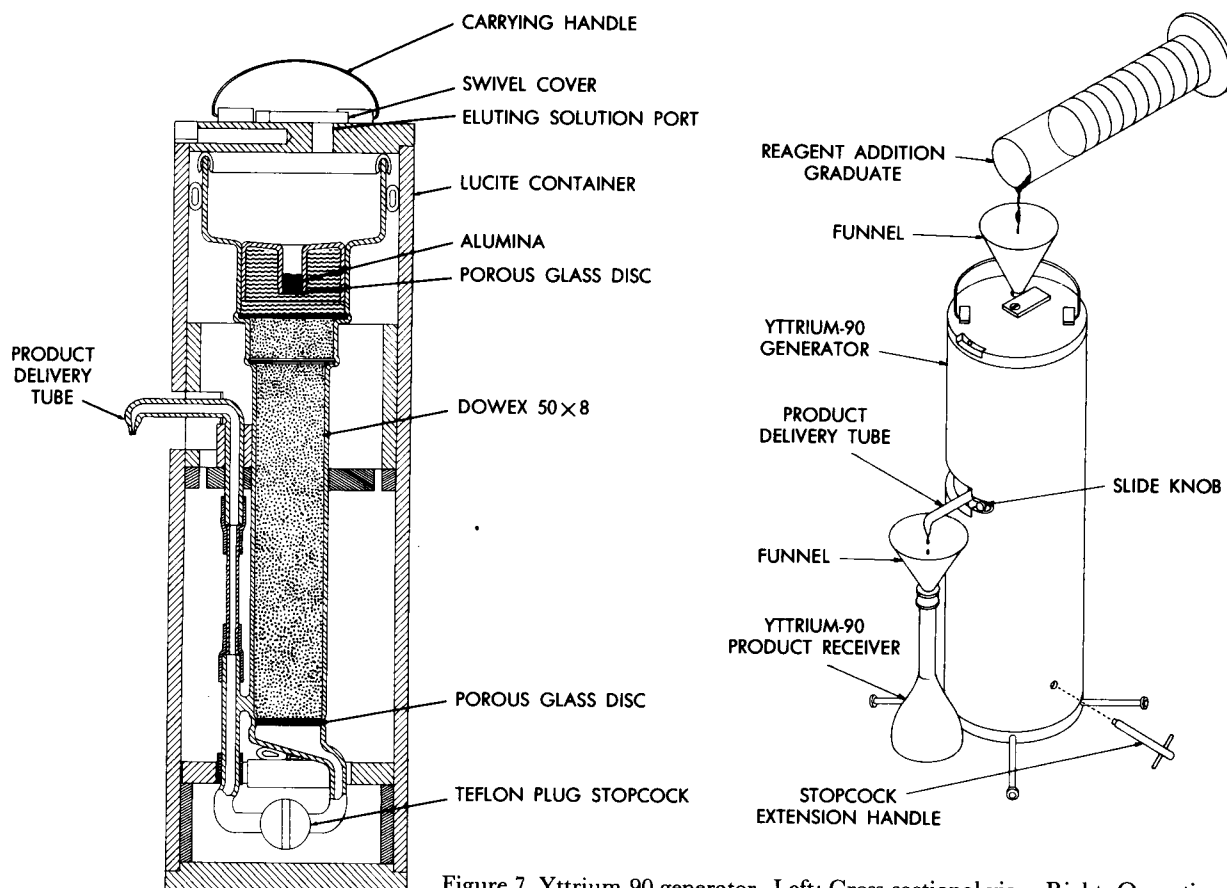


Figure 7. Yttrium-90 generator. Left: Cross-sectional view. Right: Operation.

specific study of the alpha-methylstyrene system by conventional kinetic techniques and the development of a dielectric loss apparatus for direct measurement of the polymerizing ionic species.

**Electron Spin Resonance Studies.** Irradiation of nylon produces free radicals whose nature and lifetimes are very much affected by oxygen, irradiation dose, temperature, and nylon crystallinity. A kinetic picture involving hydrogen atom migration and double bond formation is being developed.

**Radiolysis of Aromatics.** Toluene has been irradiated with various radiation doses, and the complex spectrum of radiolysis products has been analyzed by gas chromatography. It is hoped that the data derived from this and future studies will form the groundwork for a theory of radiation effects in substituted aromatic compounds.

**Radiation Catalysis.** Irradiation of MgO pretreated at 350°C produced a material of temporary high catalytic activity for *ortho-para* hydrogen conversion. No radiation effect was observed on MgO pretreated above 500°C. Studies were made of surface area, adsorption rate, and electron spin

resonance, but no fruitful correlation with catalytic effects was possible. There is a possibility that the effects were due to radiation-induced valence changes in impurities such as chromium.

#### Radioisotopes Development

**Yttrium-90.** The Brookhaven Y<sup>90</sup> generator, loaded with the long-lived parent, Sr<sup>90</sup>, will permit the 64-hr beta-emitting daughter Y<sup>90</sup> to be "milked" repeatedly as desired in the customer's own laboratory over a period of  $\approx 1$  yr. Final design of the generator (see Figure 7) and of its shipping container was completed, and several units were fabricated, the first one being sent to the Oak Ridge Institute for Nuclear Studies for field checking. Long-term tests for Sr<sup>90</sup> contamination in the product Y<sup>90</sup> solution, made on old generators at Brookhaven, continue to indicate satisfactory, safe, reliable operation during and, in some cases, beyond the 1-yr life of the generator.

The apparent nonreproducibility of the Sr<sup>90</sup> assay method has been corrected by the use of a more suitable yttrium concentration in the reagent

used in the assay. The new reagent gives a decontamination factor of  $10^7$  for  $Y^{90}$  on a single pass through the auxiliary assay column. An yttrium-citrate complex proved to be the means by which yttrium, a cation, is bound to the anion exchange resin used in this auxiliary assay column. The bicarbonate form of the Dowex 1 resin was found to be the best for assay purposes. Repeated tests have shown that a single assay column is good for at least 35 assays before a fresh column is required.

**Calcium-47.** The problem of producing  $Ca^{47}$  can be investigated by two major methods, the first proceeding from  $Ca^{46}$  as a starting material, and the second from  $Ti^{50}$ . Most, but not all, of the work done with  $Ca^{46}$  consisted of attempts to effect a Szilard-Chalmers reaction, a process in which a suitable target is chosen such that bombardment with neutrons produces recoil  $Ca^{47}$  in a form readily separable from the original target. Most of this work was devoted to the preparation and study of calcium phthalocyanine as a possible target.

Other possible production methods studied, starting with  $Ca^{46}$  but not involving the Szilard-Chalmers reaction, included spallation of a material such as  $V^{51}$  and pre-enrichment of  $Ca^{46}$  prior to Calutron separation. In the second category, four recrystallizations of  $CaNO_3$  failed to alter the  $Ca^{46}$  abundance; however, electromigration and liquid-liquid extraction remain as possibilities yet to be studied.

The second major method, starting with  $Ti^{50}$ , involves questions of nuclear physics as well as of chemical separation of any  $Ca^{47}$  produced from the original  $Ti^{50}$ . In the first category the computer was used to calculate the excitation function for the  $Ti^{50}(n,\alpha)Ca^{47}$  reaction. A maximum cross section of 4 to 8 mb at a neutron energy of  $17.5 \pm 2.0$  Mev was predicted. Attempts are being made to verify this figure experimentally. Energy losses of 14-Mev neutrons by scattering in various titanium-containing targets were also calculated.

Several satisfactory chemical methods of separating calcium from titanium with high recoveries of each were developed.  $TiF_4$  may be irradiated, sublimed away from  $Ca^{47}$ , and reirradiated. Irradiation of  $(C_5H_5)_2TiCl_2$  and extraction of  $Ca^{47}$  with HCl from a chloroform solution is also feasible. Even "dead burnt"  $TiO_2$  can be irradiated, inasmuch as it was found to be readily soluble in ammonium sulfate-sulfuric acid solution.

**Copper-67.** Pure  $Cu^{67}$  (61-hr half-life) is the most suitable radioisotope of copper for many

medical tracer experiments, including those on Wilson's disease. The  $Ni^{64}(\alpha,p)Cu^{67}$  reaction was chosen, and the yield was determined to be 0.0266  $\mu C$   $Cu^{67}$  per  $\mu amp$ -hr by using the internal alpha beam of the BNL 60-in. cyclotron. By using enriched nickel (reported to be 95%  $Ni^{64}$ ) as a target, millicurie amounts of  $Cu^{67}$  contaminated only with  $\approx 7\%$   $Cu^{64}$  have been prepared.

**Argon-38.** The production of high purity  $Ar^{38}$ , of interest as a standard for calibrating spectrographs used in geochemical and astrochemical studies, has been undertaken. The method, a modification of one used by O.A. Schaeffer (BNL Chemistry Department), consists of irradiating high purity KCl in the reactor and then introducing it into a vacuum system, where the KCl is melted and the  $Ar^{38}$  is pumped off.

**Magnesium-28.** Modifications were made in the  $Mg^{28}$  production process to remove  $H^{18}$  at an earlier stage, which permitted more accurate  $Mg^{28}$  assays, and to reduce the  $H^3$  contamination of the final product.

**Iodine-124.** Production of millicurie amounts of  $I^{124}$  from antimony targets bombarded by 40-Mev alphas in the BNL 60-in. cyclotron was shown to be feasible.

**Scandium-47.** Irradiation of calcium metal and also of CaO showed that the production of millicurie quantities of  $Sc^{47}$  is feasible in the BNL Graphite Research Reactor. A process involving carrying the desired activity on ferric hydroxide with subsequent purification by a cation exchange column and producing iron- and carrier-free  $Sc^{47}$  was developed.

**Polonium-210 (Alpha) Needle.** Fabrication of an alpha-emitting needle for use in cytological studies was undertaken. The needle is to be a right cylinder 125  $\mu$  in diameter, with 0.005  $\mu C$   $Po^{210}$  plated only on the end, the activity and the sides of the needle being covered by a chromium plate 2  $\mu$  thick. Techniques for forming the end and for measuring and controlling the thickness of the chromium cover plate were worked out; the measurement involved the projection of an electron microscope image of the needle.

**Krypton-83m.** In response to a request from Project Matterhorn (the thermonuclear research at Princeton), work was begun on the preparation of several curies of  $Kr^{83m}$  to be used at Princeton as an "artificial plasma" for testing certain aspects of the Stellarator. Purity requirements are extremely stringent, and as a result only a minimum of time



can be allowed during the necessary chemical processing step. Production will involve the irradiation of  $\approx 1$  kg high purity selenium.

**Gallium-68.** Development was begun of a simple milking system to separate from its 275-day parent,  $\text{Ge}^{68}$ , the 68-min positron-emitting daughter,  $\text{Ga}^{68}$ , which is of interest for its possible application in brain tumor localization.

**Samarium-151.** The process for separating  $\text{Sm}^{151}$  from aged fission product solution has been scaled up. The specific activity of  $\text{Sm}^{151}$  obtainable from irradiation of natural neodymium for 1 yr at a flux of  $7 \times 10^{12}$  was calculated to be 4.8 mCi/mg.

**Tellurium-132 - Iodine-132.** Yields of  $\text{Te}^{132}$  have been improved somewhat by dissolving the irradiated Al-U alloy in mercury-catalyzed  $\text{HNO}_3$ , and the amount of  $\text{Al}(\text{NO}_3)_3$  and mercury present were found to have no adverse effect on the yield. Several improvements were made in assay procedures. It was found for example that the apparent beta count of radioactive AgI is still a function of the sample thickness, even when the sample is relatively thin.

**Molybdenum-99 - Technetium-99m.** These isotopes are by-products of the  $\text{Te}^{132}$ - $\text{I}^{132}$  production process. The mercury-catalyzed  $\text{HNO}_3$  dissolution step referred to immediately above improved the  $\text{Mo}^{99}$ - $\text{Tc}^{99m}$  yields by a factor of 17.

**Fast Neutron Source.** Irradiation of pure, solid  $\text{Li}^6\text{H}^2$  showed that the ratio of 14-Mev neutrons inside the sample to thermal neutrons outside the sample was  $6.6 \times 10^{-5}$ , which is 35 to 80% of that theoretically possible (depending upon what assumptions are made in calculating the theoretical upper limit).  $\text{Li}^6\text{H}^2$  has already been put into actual use and has been found to be a more convenient source of 14-Mev neutrons than an accelerator.

## High Temperature Technology

**High Temperature Physical Property Measurements.** The goal of this study is to measure some of the physical properties of refractory nuclear fuel materials at temperatures  $> 2000^\circ\text{C}$ . For this purpose an arc image furnace to produce the high temperatures will be used, with x-ray diffraction techniques for performing the measurements. A 60-in. parabolic image furnace has been set up, and temperature measuring techniques using optical and two-color temperature meters are being tested. In addition, an inert arc for the image furnace is being developed. Preliminary x-ray measurements at temperatures up to  $900^\circ\text{C}$  have been

made with standard x-ray furnace equipment on  $\text{ThO}_2$  and several oxides of uranium.

### Release of Fission Products From Fuel Elements.

The diffusion of xenon and iodine through aluminum, stainless steel, and the Piqua fuel element cladding (0.143-in.-thick aluminum plus 0.0005 in. nickel bonding layer) has been studied with very sensitive radioactive tracer techniques. It was found that xenon does not diffuse through aluminum as thin as 0.010 in. in the temperature range  $295^\circ$  to  $473^\circ\text{C}$ . Stainless steel (type 304) 0.020 in. thick showed no evidence of diffusion up to  $610^\circ\text{C}$ . A maximum value for xenon permeability through the Piqua fuel element cladding of  $2.3 \times 10^{-11} \text{ cm}^3(\text{STP})/\text{cm}^2\text{-sec}/(\text{cm Hg/mm})$  was obtained; however, a lack of temperature correlation of the Piqua cladding results points to their anomalousness and makes them subject to suspicion.

There was no diffusion of iodine through 0.035 in. aluminum at  $461^\circ\text{C}$ .

### High Temperature Thermocouple Development.

The behavior of W-Re, W-C, and Re-C thermocouples was investigated for possible use at temperatures of  $2000^\circ\text{C}$  and above. In all cases the thermocouples were in contact with graphite. Formation of carbides of tungsten was noticed with resulting drift in the temperature-emf characteristics of the couples. The very limited experimental data available on the Re-C couple showed that in spite of its relatively low thermoelectric power (11.51 mv at  $1540^\circ\text{C}$ ) it was the most promising because of the absence of carbide formation.

### Diffusion and Absorption of Xenon in Graphite.

The diffusion of xenon in high density graphite ( $1.92 \text{ g/cm}^3$ ) at  $750^\circ$  and  $1000^\circ\text{C}$  was measured. The measured effective diffusivities were  $6.4 \times 10^{-5} \text{ cm}^2/\text{sec}$  at  $750^\circ\text{C}$  and  $9.7 \times 10^{-4} \text{ cm}^2/\text{sec}$  at  $1000^\circ\text{C}$ . The predominant mechanism by which xenon is transported through this type of graphite is flow through the interconnected pores.

Equilibrium absorption studies at the same temperatures show clearly that the xenon found in the graphite is contained in the pore volume and is not adsorbed to any measurable degree on the surfaces. This permits an accurate calculation of the quantities of xenon that may be absorbed in other graphites, provided that the volume of the interconnected pores is known from independent experiments.

## Fuel Reprocessing

### Fuel Element Reprocessing in Gas Fluidized Beds.

In the reprocessing of spent solid fuels from nucle-

ar reactors by volatilization, one of the major problems is the removal of heat from highly exothermic gas-solid reactions. It has been demonstrated that a fluidized bed of inert granular materials, because of its heat capacity and heat transfer properties, can be used to control such exothermic reactions. Thus, the fluidized bed is used in the same way that liquids in the form of molten salts, interhalogens, etc., are used, without the introduction of problems encountered in the use of such liquids.

As outlined in earlier reports on the development of the fluidized bed process, it was anticipated that the process would have wide application in the field of reprocessing irradiated fuels. Subsequent experiments have shown that it is possible to apply this method to fuels consisting of U metal,  $\text{UO}_2$ , or UC, and to fuels alloyed or clad with Zircaloy. For these fuels the following reactions have been demonstrated to be practical.

- 1) UC:  $3\text{UC} + 7\text{O}_2 \rightarrow \text{U}_3\text{O}_8 + 3\text{CO}_2$ ;  
 $\text{U}_3\text{O}_8 + 9\text{F}_2 \rightarrow 3\text{UF}_6 + 4\text{O}_2$ .
- 2)  $\text{UO}_2$ :  $\text{UO}_2 + 3\text{F}_2 \rightarrow \text{UF}_6 + \text{O}_2$ .
- 3) U metal:  $\text{U} + 3\text{F}_2 \rightarrow \text{UF}_6$ .
- 4) Zircaloy-clad or alloyed fuels:  
 $\text{Zr} + 4\text{HCl} \rightarrow \text{ZrCl}_4 + 2\text{H}_2$ ,  
 followed by fluorination of U.

For the past year the above reactions have been studied in detail with emphasis on uranium recovery and the hydrochlorination of elements of the Zircaloy type. In the case of fuels of the Zircaloy type, hydrochlorination of 10-in. sections of multiplate elements has been routinely carried out in a 6-in.-diam hydrochlorinator with negligible uranium loss. It has also been established that  $\text{Al}_2\text{O}_3$  (Reg. type, Norton Co.) is suitable as an inert bed.

From the above considerations it is concluded that the reprocessing of nuclear fuel elements via the inert fluidized bed method can be applied to any fuel in which the major components can be selectively volatilized, regardless of the exothermic nature of any particular reaction.

**Fluoride Volatility Process.** The liquid mixture  $\text{NO}_2$ -HF has proved to be a remarkably versatile reagent for most of the materials used in the fuel elements of existing or proposed power reactors. It attacks uranium, zirconium, Zircaloy, stainless steel, aluminum, niobium, molybdenum, beryllium, and uranium oxide, and converts the metals

into complex fluorides containing oxides of nitrogen. These can be decomposed to the normal fluorides by moderate heating. For a "dissolution" or reaction step to be suitable for incorporation into a fuel processing cycle, the solid should react at a rate  $>5 \text{ mg/cm}^2\text{-min}$ . For the materials listed, this rate is achieved in  $\text{NO}_2$ -HF at temperatures of  $100^\circ$  to  $150^\circ\text{C}$ . The corresponding pressures are 100 to 350 psig, for which it should not be difficult to design adequate containment. The best material of construction for the reaction vessel appears to be Monel, which corrosion tests have shown to stand up well under the conditions used in the reaction. A complete fuel processing scheme using the new reagent has been worked out and called the nitrofluor process. It involves the following steps, in the particular case of a high zirconium Naval reactor fuel element: (1) reaction with 75 M % HF - 25 M %  $\text{NO}_2$ ; (2) separation of liquid phase from solid zirconium fluoride; (3) evaporation to dryness of liquid phase with recovery of reagent; (4) heating of the uranium-containing residue from the evaporation step to produce normal  $\text{UF}_4$ ; (5) treatment with  $\text{BrF}_3$  to convert  $\text{UF}_4$  to  $\text{UF}_6$ ; and (6) fractional distillation to purify  $\text{UF}_6$  from all other volatile components.

The research program on the circumstances leading to ignition and explosion in U- $\text{BrF}_3$  systems was completed. It was concluded that the dissolution of uranium metal in any reagent containing  $\text{BrF}_3$  is potentially hazardous.

**Aqueous Fuel Reprocessing.** Process development work was initiated on aqueous methods for the recovery of uranium from the more advanced designs of Naval reactor fuel. Work has been carried through the "cold" pilot-plant stage for two processes: nitric-hydrofluoric acid and ammonium fluoride-ammonium nitrate. Uranium can be recovered from the resulting solutions by a modified flow sheet of the Purex type.

An interim summary report entitled *Aqueous Processes for Naval Reactor Core* is in preparation and will be issued as a classified report.

**Amalgam Reduction of  $\text{UO}_2$ .** Reaction rate studies on uranium oxide-calcium amalgam are being conducted in a newly built walk-in hood. No reduction was obtained at  $300^\circ\text{C}$  for high fired  $\text{UO}_2$  spheres after 40 days or for sulfated feed material  $\text{UO}_3$  over 9 days. (Previous positive results may have resulted from faulty sampling techniques.) Future effort will be directed toward long-term equilibrium studies in a salt-heated rotating

sampler, and an attempt will be made to determine whether an induction period exists.

An inert atmosphere dry box has been constructed for experiments on galvanic cell activity measurements and for gas – amalgam kinetic reactions.

#### **Preparation of Tetravalent Uranium Compounds.**

Several methods have been investigated for the preparation of U(IV) solutions that can be used for reduction of plutonium in the Purex process partition cycle. One method includes solution of  $\text{UO}_2$  (prepared by hydrogen reduction at  $600^\circ\text{C}$ ) in various  $\text{H}_2\text{SO}_4$ –HF solutions, separation from sulfate ion by precipitation of the uranium hydroxide with  $\text{NH}_3$ , and dissolution into  $\text{HNO}_3$ – $\text{NH}_2\text{SO}_3\text{H}$  mixtures. In a second method  $(\text{NH}_4)_2\text{S}$  is used to precipitate  $\text{UO}_2\text{S}$  from  $\text{UO}_2(\text{NO}_3)_2$  solution. Digestion of the  $\text{UO}_2\text{S}$  with excess  $(\text{NH}_4)_2\text{S}$  yields a  $\text{UO}_2$  that is readily soluble in  $\text{HNO}_3$ – $\text{NH}_2\text{SO}_3\text{H}$  or  $\text{HNO}_3$ – $\text{NH}_2\text{SO}_3\text{H}$ –HF mixtures. Other less favorable methods centered on precipitation of uranium from nitrate solution with organic amine compounds, followed by either calcination in an inert atmosphere at elevated temperature to produce  $\text{UO}_2$ , or dissolution and boiling with concentrated  $\text{H}_2\text{SO}_4$  to yield U(IV).

### **Waste Chemical Technology**

#### **Waste Treatment (Long-Lived Nuclide Removal).**

Experiments have been conducted on the concentration of long-lived fission products (cesium, strontium, rare earth) and transuranium elements from Purex and TBP-25 process wastes by using solvent extraction techniques. The long-lived fission products can be extracted from carefully adjusted iodide solutions by mixtures of iodine in nitrobenzene and back-extracted by  $\text{HNO}_3$  solution. Extraction coefficients have been determined in batch and mixer-settler experiments using varying concentrations of fission product ions, extractant compositions, and feed solutions.

**Xenon Adsorption on Activated Carbon.** The continuous adsorption of xenon from a helium stream on activated carbon (Columbia HCC 12/28 mesh and cxc 6/8 mesh) has been studied in order to develop information necessary for the design of purification systems for gas coolants. The experiments were carried out in a 1.38-in.-i.d. column, 10.5 in. long, containing 141 g carbon. The important variables studied and the conclusions as to their effect are as follows.

1) Temperature in the range  $25^\circ$  to  $100^\circ\text{C}$ . Its effect is shown to be due to the variation with tem-

perature of the equilibrium adsorption isotherm and of the diffusivity.

2) Concentration of xenon in helium in the range 0.050 to 1.31%. As expected from the isotherm relationship, increased partial pressure of xenon in the gas stream results in higher adsorptions per unit volume of carbon. An increase in the slope of the adsorption wave is also noted with increasing xenon concentration.

3) Mass velocity in the range 10 to 65 lb/hr-ft<sup>2</sup>. The performance of the adsorber bed improves considerably as the flow rate is increased to 40 lb/hr-ft<sup>2</sup> and then drops off substantially because of channeling and other "overloading" effects.

Comparison of these results with the theory of the general type of unsteady-state processes involved in fixed bed adsorption shows good agreement.

**Ultimate Waste Disposal.** The adsorption and fixation of fission products on mineral exchange material has been shown to be a very effective method of disposing of long-lived radioactive isotopes, notably  $\text{Cs}^{137}$  and  $\text{Sr}^{90}$ . The ion exchange process lends itself to practical operation, provided that corrosion products and other bulk ions are not present in excessive concentrations. Experiments over the past year have centered on the investigation of methods of neutralizing the effects of these interfering ions. Chelation of the corrosion products with citric acid or acid pretreatment of the mineral exchanger to convert it to the hydrogen form has made it possible to obtain decontamination factors with respect to strontium of the order of  $10^9$ .

The net result of this work, which has been in progress for more than ten years, is that permanent disposal of the long-lived fission product wastes can readily be obtained by the adsorption and fixation on mineral ion exchange materials.

Another method of converting liquid wastes to the stable solid form, developed at Brookhaven, is the calcination of wastes containing high concentrations of metal salts to produce the stable oxides such as  $\text{Al}_2\text{O}_3$  and  $\text{ZrO}_2$ .

Since calcination is simply a thermal decomposition step, a number of mechanical devices may be used to carry out this operation. One such device is the rotary ball kiln which has been used throughout the BNL studies. During the past year, work has centered on construction of a small pilot plant. The calciner, with a capacity of 5 gal/hr of liquid feed, is 8 in. in diam and has a heated length of 7 ft.

Since a very high degree of control of radioactive dust in the off-gases from the calcination is imperative, the rotary calciner has the advantage that the off-gases are almost entirely condensable.

### Radiation Engineering

Theoretical and experimental comparisons have been made of large-scale, low level slab irradiators using  $\text{Na}^{24}$ ,  $\text{Cs}^{137}$ , and  $\text{Co}^{60}$ , and of  $\text{Cs}^{137}$  slab sources.

Shielding experiments were performed to determine the radiation scattered from various duct and  $\text{Co}^{60}$  source arrangements that may be encountered in future high level radiation facilities.

Gamma radiation facilities were designed and built for the AEC exhibits in New Delhi, India, and Cairo, Egypt. These were installed and operated by BNL personnel.

A gamma facility employing  $\approx 25,000$  C of  $\text{Co}^{60}$  has been designed and is being built. This first of a number of units to be built will be installed at the Massachusetts Institute of Technology for use in the Food Irradiation Research program.

Work with AMF Atomics on the design of the high level source development laboratory cell and canal complex is about completed, and construction will begin in fiscal 1961.

### Radiation Processing With Chemonuclear Reactors

The object of this program is to investigate the economic feasibility of using radiation energy for the production of valuable chemicals. Two extensive engineering evaluation studies on chemonuclear processes, including a thorough review of the utilization of fission fragment energy for the fixation of nitrogen, have been completed. These studies cover radiation chemistry, fuel problems, reactor design, chemical process design, and economic evaluation. From these paper studies it was concluded that there is no inherent technical barrier against the large-scale production of fixed nitrogen, but that under the present optimistic conditions, even in a nuclear economy, fission recoil energy presents no economic advantages over other indirect processes.

In the experimental program, chemical yields of  $\text{NO}_2$  in the nitrogen fixation reaction have been determined at high temperature and pressure by using cobalt gamma radiation, and significant solid wall effects were noted. Preliminary data were

also obtained on the yield of hydrazine from irradiation of aqueous ammonia solutions.

### Liquid Metal Heat Transfer Research

Nearly all the liquid metal heat transfer research conducted at BNL in recent years has been concerned with flow of liquid metals through tube banks. Two related analytical studies completed during the year resulted in equations for determining the heat transfer coefficients for the fully established, turbulent, and parallel flow of liquid metals through unbaffled tube banks. These equations are

$$\text{Nu} = 0.93 + (10.81 P/D) - 2.01(P/D)^2 + 0.0252(P/D)^{0.273}(\psi \text{Pe})^{0.8} \quad (1)$$

and

$$\text{Nu} = 7.0 + 3.8(P/D)^{1.52} + 0.27(P/D)^{0.27}(\psi \text{Pe})^{0.8}, \quad (2)$$

where

$\text{Nu}$  = Nusselt number  $= hD_e/k$ ,

$h$  = average heat transfer coefficient,

$D_e$  = equivalent diameter of tube bundle,

$k$  = thermal conductivity of liquid metal,

$P$  = distance between tube center, equilateral triangular spacing,

$D$  = outside diameter of tubes,

$\psi = \epsilon_H/\epsilon_M$ ,

$\epsilon_H$  = eddy diffusivity for heat transfer,

$\epsilon_M$  = eddy diffusivity for momentum transfer,

$\text{Pe}$  = Peclet number  $= D_e v_a C_p \rho / k$ ,

$v_a$  = average linear velocity,

$\rho$  = density of liquid metal, and

$C_p$  = heat capacity of liquid metal.

Depending upon conditions, Equation (2) gives coefficients that are 10 to 20% below those given by Equation (1). The methods of deriving the two equations differed principally in two ways: mathematical treatment, and the velocity distribution data used. Equation (1) is based upon the most recent and presumably more accurate data.

Experimental results were obtained on a mercury loop, which may be compared with those predicted by the above equations. At a Peclet number of 500, the experimental curve (from averaged results) fell 27% below that predicted by Equation (1) and 33% below that predicted by Equation (2), while at a Peclet number of 2000, the corresponding deviations were 7 and 12%. The comparisons were made on the basis of  $\psi = 1.00$ .

The above results are the first to be published for this particular system. The discrepancy between theory and experiment, typical of liquid metal systems, cannot yet be satisfactorily explained. It is hoped that further experiments with improved equipment will yield results of greater accuracy and reproducibility, to establish more firmly the experimental aspect of the study.

Some work was also carried out on the analytical determination of heat transfer coefficients for liquid metals flowing in annuli.

### Reactor Component Development

**Fluorocarbons.** The fluorocarbons form a class of compounds coextensive in theory with hydrocarbons and similar to them. If fluorocarbons in general (or certain ones) should also prove to have good stability in ionizing radiation, they might well find application in nuclear technology as coolants, slurry vehicles, hydraulic fluids, etc. A program of synthesis and testing of promising compounds has therefore been initiated. Emphasis is being placed on the fluorocarbon analogues of the hydrocarbons that have proved to be most resistant to radiation: naphthalene, biphenyl, and the terphenyls.

**Gas Coolants.** The purpose of this project is the development of gas coolants suitable for use in the various reactor-steam turbine and reactor-gas turbine systems under development. The coolants now used are helium,  $\text{CO}_2$ , and nitrogen, which are not completely satisfactory. The principal qualities desired in a good gas coolant are (1) good heat transport and transfer capability, (2) low pumping power requirement, (3) resistance to decomposition, and (4) high molecular weight.

A thorough study of gases from the standpoint of thermodynamic performance has been initiated. Coupled with this, an experimental testing program for thermal resistance and effect of container materials has been started. As certain gases show promise in these tests, they will be subjected to radiation decomposition tests.

From the data obtained so far, a mixture of gases will probably give the best set of properties for a coolant. This will allow combining the good properties of various gases. The gases under consideration are helium, hydrogen, nitrogen,  $\text{CO}$ ,  $\text{CO}_2$ , argon, neon,  $\text{CF}_4$ ,  $\text{SF}_6$ ,  $\text{C}_2\text{F}_6$ ,  $\text{C}_3\text{F}_8$ ,  $\text{C}_4\text{F}_{10}$ ,  $\text{CS}_2$ ,  $\text{COS}$ ,  $\text{SO}_2$ ,  $\text{C}_6\text{H}_5\text{F}$ ,  $\text{C}_6\text{H}_6$ ,  $\text{CCl}_2\text{F}_2$ , and  $\text{CHClF}_4$ . Typical gas mixtures are  $\text{CF}_4$  and hydro-

gen;  $\text{CF}_4$  and helium;  $\text{CF}_4$ , helium, and hydrogen; and  $\text{CO}_2$  and hydrogen.

### LIQUID METAL FUEL REACTOR

In the spring of 1959 the AEC decided to discontinue the Liquid Metal Fuel Reactor Experiment (LMFRE) as such. The program for fiscal 1960 has two aims:

1. Close out the LMFRE, which included operation of the Utility Test Loop, the Radiation Loop, Loop G, and the High Velocity Loops;
2. Continue certain basic work in liquid metal fuel technology, including slurries, materials, and some chemical processing.

As of June 1960, all this work has been completed except for that with the Radiation Loop, which is still in operation in the Brookhaven Graphite Research Reactor. The satisfactory operation of these large loops has reinforced our confidence in the LMFR concept.

### Engineering Development

**Utility Test Loop.** The 4-in. Utility Test Loop was built to provide information on the design, construction, and operation of large liquid bismuth fuel systems. The loop was designed to operate at a power level of 1.8 Mw, with a flow rate of 360 gal/min in each loop. Under  $\Delta T$  conditions the furnace outlet temperature was maintained at 894°F, and the cooler outlet temperature at 750°F.

In October 1959, Brookhaven completed the initial loop circulating tests which included the addition of magnesium and zirconium to the charge, piping conditioning runs, and a 48-hr run at design conditions. During this initial operating period some component malfunctions occurred, the failure of the circulating pumps being the most serious. Suitable repairs were made and no further trouble was experienced with these components.

With the completion of the 48-hr  $\Delta T$  run the responsibility for the loop was transferred to the Babcock & Wilcox Company, who then conducted performance tests on the loop and its components. Both steady-state and transient characteristics of the equipment were determined. The results of these tests are presented in detail in *Liquid Metal Fuel Reactor Experiment, Operation and Testing of a Nonnuclear Liquid Metal System*, BAW-1192, May 1960. This phase of the experiment was completed on January 4, 1960, and the loop was shut down with the termination of the LMFR program. BNL

then assumed the responsibility for disassembly and inspection of the loop.

The loop was operated for a total of 1700 hr, 800 hr under  $\Delta T$  conditions ( $\Delta T = 75^\circ$  to  $100^\circ\text{F}$ ) at a power level of 1.2 Mw, and 900 hr under isothermal conditions at  $1000^\circ\text{F}$ . A short ( $< 1$  hr) run at 4 Mw was made to determine the maximum capability of the system.

The results of the experiment were very encouraging with respect to the application of small loop techniques and specifications to large-scale equipment. Additive stability was excellent. Only slight corrosion was observed in the furnace and metallurgical specimen sections of the loop. Some corrosive attack was noted in the bismuth circulating pumps. A detailed presentation of the results of the experiment based on the final examination of the loop will be published as a BNL report.

**Radiation Loop.** The Radiation Loop, designed to study the effect of radiation on the corrosion of low chrome steels, carbon steel, molybdenum, and beryllium by a circulating U-Bi fluid, was installed in the Brookhaven Graphite Research Reactor in April 1960. A series of runs, during which the loop shielding and thermal balance were checked, were made at increasing concentrations of enriched uranium. The final concentration of uranium in bismuth (975 ppm  $\text{U}^{235}$ ) was reached, and the specified temperature differential ( $500^\circ$  to  $425^\circ\text{C}$ ) was established on June 9, 1960. At the end of the fiscal year, 394 hr of in-pile operation under these conditions were completed. The total in-pile operating time under all conditions was 557 hr. The loop operating conditions are as follows:

1. Fluid: Bismuth containing 975 ppm  $\text{U}^{235}$ , 240 ppm Zr, and 350 ppm Mg;
2. Flow rate: 5 gal/min;
3. Temperature: In-pile tip,  $500^\circ\text{C}$ ; cooler exit,  $425^\circ\text{C}$ .

Prior to the in-pile run, a 500-hr out-of-pile run was made under similar conditions. It is planned to continue the run until at least 3000 hr of in-pile operation have been completed.

Numerous in-pile capsule tests were run to study the effectiveness of zirconium in inhibiting corrosion of carbon and low alloy steels in fissioning U-Bi solutions. The results of this investigation indicate that corrosion was considerably enhanced by the neutron and fission fragment bombardment. Materials tested were chrome steels, carbon steels, molybdenum, and tantalum; all except the

last were severely attacked at  $500^\circ$  to  $600^\circ\text{C}$  in 1000 to 2000 hr.

The High Level Metallurgical Hot Cell has been completed and is now ready to receive the Radiation Loop.

**Loop G.** As a control on the Radiation Loop, Loop G was operated under identical conditions, but without radiation, for 3000 hr and was shut down for examination in October 1959.

**High Velocity and Thermal Convection Loops.** During the year  $\approx 95\%$  of the loop work connected with the LMFR program was discontinued. The remaining loop work consisted of operating seven thermal convection loops in order to study the effect of U-Bi plus Zr and Mg on the corrosion and mass transfer of carbon steel over an extended period of time (10,000 hr). Operation of the thermal convection and pumped loops has led to the following new conclusions on the corrosion and mass transfer behavior of steels in contact with U-Bi.

1. Carbon steels are the least susceptible to attack of all the steels tested. However, increasing the temperature differential above  $150^\circ\text{C}$  and the maximum temperature to  $650^\circ\text{C}$  causes detectable corrosion of carbon steels after  $\approx 5000$  hr in a thermal convection loop.

2. Increasing the liquid metal velocity to  $\approx 10$  ft/sec rapidly increases the rate of attack on  $2\frac{1}{4}$  Cr - 1 Mo steels. A pumped loop was operated for 190 hr at a  $550^\circ$  to  $400^\circ\text{C}$   $\Delta T$  and 10 ft/sec, at which time the  $2\frac{1}{4}$  Cr - 1 Mo pipe wall (0.109 in. thick) was corroded through. Immediately adjacent pipe made of  $1\frac{1}{4}$  Cr -  $\frac{1}{2}$  Mo steel was not attacked. Attack was most severe where surface irregularities disturbed the flow pattern.

3. Increasing the liquid metal velocity to 10 ft/sec and operating at a  $550^\circ$  to  $400^\circ\text{C}$   $\Delta T$  did not cause detectable corrosion of carbon steel specimens after 640 hr.

4. Several loops made of coextruded pipe, carbon steel inside and  $1\frac{1}{4}$  Cr -  $\frac{1}{2}$  Mo steel outside, have operated for up to 10,000 hr at a  $125^\circ\text{C}$   $\Delta T$  with no detectable corrosion.

5. 1000 ppm uranium can be maintained in a mechanically pumped loop for  $> 6000$  hr.

### Technology (Fuel and Blanket Development)

**ThBi<sub>2</sub>-Bi Slurries.** Study of the possible use of a slurry of solid ThBi<sub>2</sub> particles in liquid bismuth as a fluid breeder-blanket material was continued, with primary emphasis on problems associated

with circulating the slurry through a temperature differential. In a slurry containing 4.8% Th, 0.4% Te, and 250 ppm Zr, increasing the ThBi<sub>2</sub> particle size by heat treatment at 800°C increased the length of time during which the slurry could be circulated in a 2¼ Cr - 1 Mo steel loop through bulk temperature differentials of 21° to 40°C (510°C max) at 2 ft/sec without deposition in the cooled section. After periods of deposit-free circulation ranging from 70 to 185 hr, deposits of viscous concentrated slurry containing ThBi<sub>2</sub> particles smaller than those initially present in the slurry were formed. Apparently, the ThBi<sub>2</sub> particles were being fractured by the pump impeller, which decreased the ThBi<sub>2</sub> particle size and increased the average viscosity of the slurry and the critical erosion velocity. Both these factors should contribute to the formation of the concentrated slurry deposits. Continued circulation after these deposits were formed led eventually to deposition of large columnar grains of ThBi<sub>2</sub> in the cooled section of the loop, which indicated that thorium was being deposited from solution. After loop shutdown, no erosion or corrosion of the loop or the pump or of graphite inserts in the loop by the circulating slurry could be detected.

In concurrent capsule-scale experiments, the mechanism by which minor additions of tellurium to ThBi<sub>2</sub>-Bi dispersions inhibit ThBi<sub>2</sub> particle growth and prevent deposition of thorium from solution during temperature differential circulation was shown to be the formation on the surface of the ThBi<sub>2</sub> particles of a thin layer of an intermetallic compound that is virtually insoluble in liquid bismuth at temperatures up to at least 800°C.

**ThO<sub>2</sub>-Bi Slurries.** Study was continued of the preparation of dispersions of ThO<sub>2</sub> in liquid bismuth for possible use as a fluid breeder-blanket material. An attempt was made to determine the mechanism by which additive elements such as magnesium and zirconium dissolved in bismuth promote the wetting and dispersion of ThO<sub>2</sub> powders. Sessile drop experiments on fused single crystals of ThO<sub>2</sub> indicated that random polished planes of the crystals could be wetted by pure liquid bismuth under vacuum at 600°C, but that cleavage planes of the crystals, (111) in all cases, could not be wetted unless the bismuth contained appreciable concentrations of magnesium or zirconium. Increasing the additive concentration in the bismuth appeared to cause lower receding contact angles on the random planes; however, wet-

ting of the cleavage planes was not improved by increasing the additive concentration beyond that required to achieve wetting.

Capsule-scale experiments indicate that 600°C-outgassed ThO<sub>2</sub> powders in which the particles are not bounded by cleavage surfaces can be dispersed in liquid bismuth containing no additives at 600°C, given sufficient agitation. Appreciable amounts of gas are evolved from the ThO<sub>2</sub> during dispersion. Other investigators suggest that this gas is primarily CO and CO<sub>2</sub> which remains chemisorbed on the ThO<sub>2</sub> surface after outgassing at 600°C. Part of the evolved gas remains trapped in the slurry, which results in porosity and pockets of unwetted ThO<sub>2</sub> powder. Above some minimum ThO<sub>2</sub> concentration, the amount of trapped gas is a function of the geometry of the system and the duration and violence of agitation, and does not vary appreciably with the ThO<sub>2</sub> concentration or surface area.

#### **Engineering-Scale Studies of ThO<sub>2</sub>-Bi Slurries.**

The effort to prepare and characterize special ThO<sub>2</sub> powder as well as kilogram-scale batches of ThO<sub>2</sub>-Bi slurries was continued, and experiments were designed and carried out to provide qualitative information on the problem of the diffusion of protactinium from ThO<sub>2</sub> particles and its adsorption thereon.

Work continued on the preparation of pure ThO<sub>2</sub> by precipitation of the hydroxide from an ether solution of Th(NO<sub>3</sub>)<sub>4</sub>, followed by calcination of the hydroxide to a free-flowing powder which was readily dispersed in pure bismuth. The characteristics of this oxide were measured.

In the engineering-scale studies, it was possible to disperse ThO<sub>2</sub> powders in concentrations up to 11.5% completely and uniformly in bismuth at 600°C and under  $\approx 5 \mu$  Hg pressure. Seven separate 50-lb batches of slurry were prepared, five containing originally 350 ppm each of Mg and Zr, the sixth containing initially 450 ppm Mg and 600 ppm Zr, and the last, 350 ppm Zr only. Vigorous agitation was required for dispersion. Commercially available oxide powder, prepared from thorium oxalate, was used in all cases except one, where ThO<sub>2</sub> prepared at Brookhaven from Th(OH)<sub>4</sub> was used. The calcination temperature, particle size range, and surface area of the oxides varied widely without apparent effect on the dispersibility of the slurry.

The ThO<sub>2</sub>-Bi slurries were found to be very stable. As expected, settling rates were very low,



and the dispersions remained uniform after standing for periods of up to 114 hr at 600°C. No temperature effect on slurry stability was found in the range of 400° to 600°C. The presence of up to 450 ppm Mg and 650 ppm Zr did not reduce the ThO<sub>2</sub> to the metal in the temperature range of 400° to 600°C.

Gases entrained in the slurry decreased its stability, with resultant nonuniformity. The major cause of instability was the formation of froth, into which the ThO<sub>2</sub> concentrated, upon reduction of the gas pressure over the liquid surface. The amount of entrained gas was found to increase with increasing agitator speed, gas pressure, and period of agitation. It was further found that surface turbulence was the most important factor in controlling gas entrainment, and that the risk of forming a froth could actually be eliminated by maintaining a quiescent surface.

**Slurry Loop.** Slurries with ThO<sub>2</sub> concentrations up to 7.4% were successfully circulated for 1900 hr at a flow rate of 1.3 gal/min (1.4 ft/sec) in a ½-in. IPS loop, under isothermal conditions at 525°C for 1000 hr, and the remainder of the time at a 30°C bulk temperature difference (515° to 485°C). In general, good slurry stability was observed, with no frothing. No corrosion or erosion was noted on a metallurgical test piece in this loop. The techniques used for circulating bismuth solutions are generally applicable to ThO<sub>2</sub>-Bi slurries if gas entrainment is prevented.

Agitator impellers ran in the slurry vessels at 600°C and at speeds of ≈200 rpm (tip speed of 17.5 ft/sec) for operating periods as long as 500 hr. These impellers, made of 2¼ Cr - 1 Mo alloy steel, showed only slight erosion. However, results from another study showed that erosion is a problem in this slurry system. In this study, anchored 2¼ Cr - 1 Mo alloy steel tabs were rotated at 500°C for 1000 hr at a tip speed of 7.5 ft/sec relative to stationary slurry in glass doughnuts. In the other phase of this work, flat 1¼ Cr - ½ Mo and 2¼ Cr - 1 Mo alloy steel tubes, in the shape of sharp-edged parallelograms, were rotated at tip speeds between 17.5 and 60 ft/sec in several slurry vessels at 500°C for periods up to 230 hr. On the basis of these tests, no unusual effects due to the presence of thorium in the bismuth were observed at 7.5 ft/sec. However, it was apparent that, at tip speeds of 17.5 ft/sec and over, erosion occurred and increased as the tip speed increased and as the concentration of additives in the bismuth (liquid)

phase decreased. It also appeared that 1¼ Cr - ½ Mo alloy steel was more resistant to erosion than 2¼ Cr - 1 Mo alloy steel.

**Protactinium Removal.** Capsule experiments were carried out to determine qualitatively the influence of temperature and time on the rate of diffusion of Pa<sup>233</sup> out of irradiated ThO<sub>2</sub> dispersed in bismuth. The results indicated that diffusion probably did not occur to any noticeable extent.

**Polonium Release From Bismuth.** The release of Po<sup>210</sup> from irradiated bismuth under 30 to 100 μ He pressure has been measured in the temperature range of 200° to 430°C. With a Po<sup>210</sup> concentration of  $2.5 \times 10^{14}$  atoms per gram, the measured releases were  $9.80 \times 10^8$  atoms/cm<sup>2</sup>-min at 200°C (solid Bi),  $1.43 \times 10^{10}$  atoms/cm<sup>2</sup>-min at 330°C (m.p. of Bi is 272°C),  $1.20 \times 10^{12}$  atoms/cm<sup>2</sup>-min at 390°C, and  $3.8 \times 10^{12}$  atoms/cm<sup>2</sup>-min at 430°C. The results show that the rate of Po<sup>210</sup> evaporation from solid bismuth is ≈1% of the rate of release when the bismuth is slightly above its melting point, and 0.1% of the rate from bismuth at ≈4000°C.

**Desorption of Xenon From U-Bi Fuel.** The removal of xenon from U-Bi fuel at 500°C was investigated in a perforated plate column and in a packed column. The former was 1.61 in. i.d. and contained 6 plates perforated with 19 ⅛-in. holes, the plates being located 3 in. apart. The packed column was 1.61 in. i.d. and 16 in. long, and was filled with ¼-in.-diam stainless steel balls. A sparging helium stream of 80 cc/min was used in all experiments. The bismuth throughput rates were 88,400 lb/hr-ft<sup>2</sup> for the packed column, and from 80,000 to 150,000 lb/hr-ft<sup>2</sup> for the perforated plate tower. There was evidence of flooding, however, at the higher rates, and a throughput of ≈100,000 lb/hr-ft<sup>2</sup> is probably the optimum for both columns. The results show that there was no difference between the two columns in efficiency for xenon removal. At least 80% of the xenon was removed in a single passage through either column.

**Distribution of Technetium.** Of the noble fission products, one of the most important from the point of view of neutron economy is technetium. Milligram quantities of Tc<sup>99</sup> became available during the year, and Bi-Tc solutions were made up and subjected to extraction experiments with both fused salts and liquid zinc. It was found that neither a strongly oxidizing salt mixture nor liquid zinc extracted more than 10 to 20% of the technetium in a single equilibration.

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 biological effects of radiation and the use of nuclear techniques, especially isotopes, for the elucidation of some of the basic problems of biology.

In general these problems are being investigated by the methods of molecular biology. This approach has been under development for several years, and the growth of the Department has been increasingly in this direction. It has involved studies on protein structure, enzyme kinetics, the molecular structure and function of antibodies, the molecular structure of chromosomes and its relation to the mutation process, etc. These concepts and techniques have found immediate application in such pressing problems as the nature of radiation-induced mutations, the intimate nature of the mechanism of radiation damage in mice, and the details of the basic reactions involved in photosynthesis. The molecular approach thus permeates the work of the Department.

This report briefly indicates the major areas of research and some of the findings made during the year.

## MAMMALIAN PHYSIOLOGY

### Radiation-Induced Aging

The decrease in life expectancy of animals caused by radiation has been called radiation-induced aging because the process resembles natural aging. Both kinds of aging are being studied in order to establish the mechanisms involved and to determine the relationship between the two processes. It has been postulated that aging results from the accumulative effects of nonspecific stresses. Tests of this possibility in mice subjected to a wide variety of stresses (e.g., tetanus toxin) in single or periodic doses have indicated that stress *per se* does not cause a decrease in life expectancy. Thus, radiation appears to differ from other stresses in that it uniquely shortens life. It is still possible that radiation constitutes a specific stress on some organ as yet unsuspected. This possibility is being

explored by consideration of the effect on life span of specific stresses, e.g., the kidney damage produced by mercury.

It has also been suggested that aging results from the accumulation of viable mutations in the somatic cells of the body. Since radiation is a mutagenic agent, the phenomenon of radiation-induced aging supports this hypothesis. However, nitrogen mustard, a chemical mutagen, was not found to shorten the life span. Although this argues against the mutation hypothesis, proof requires quantitative estimates of the mutations produced by the chemical and by the radiation. Such estimates have now been made for liver, based on the following considerations: Mutations may be assumed to be proportional to chromosome aberrations. If an abnormality is produced in a chromosome during interphase, it will not become apparent until cell division occurs. Since liver cells divide infrequently, chromosome abnormalities can be "stored" for long periods of time. However, destruction of part of the liver induces the remaining cells to divide. Determination of the proportion of the dividing cells that exhibit abnormalities then gives an estimate of somatic mutations.

Results to date indicate that the number of chromosome abnormalities increases linearly as the animal grows older. The increase is somewhat greater in animals given periodic doses of nitrogen mustard than in controls. In contrast, radiation markedly increases the number of abnormalities. It seems justifiable to conclude tentatively that somatic mutations do play an important part in the natural aging process. In addition, the small increase in mutations produced by nitrogen mustard predicts the observed negligible effects on life span. The marked increase in mutations produced by radiation strengthens the somatic mutation theory of aging.

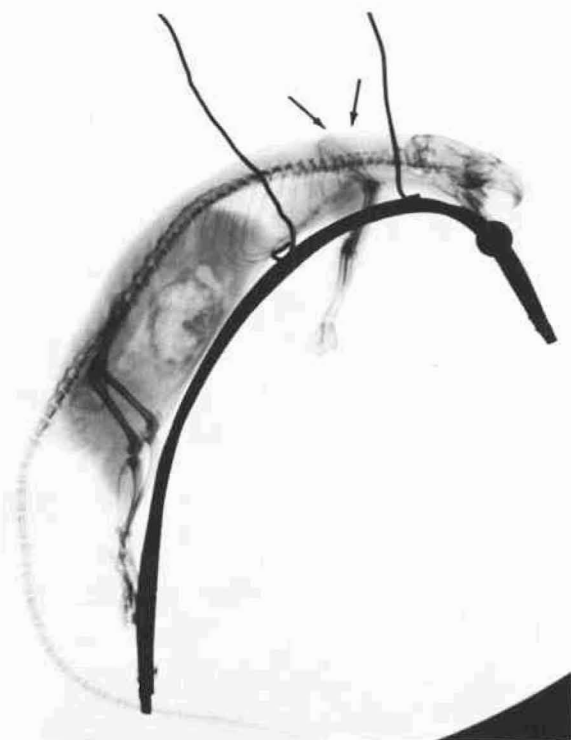
### Physiological Effects of Irradiation

Rhythmical weight variations occurring in female rats have been correlated with cyclic estral activity. The animals gain weight rapidly during the first two days of their four-day cycle. This gain

ceases on the third day and gives way to a small loss on the fourth day. The whole-body application of 600 r of x-radiation produces a sharp weight loss for three days postirradiation. Although the animals show no cyclic weight patterns during this period, they do maintain vaginal indications of estral cycling. This level of x-radiation does not seem to alter estrogenic activity.

The liver glycogen and blood sugar of normal female rats reach maximum and minimum values, respectively, on the second day of estrus. By the fourth day the situation is reversed. Fasting imposed at any point in estrus causes a drop in liver glycogen and blood sugar to identical base values. Irradiated, fasted females showed higher liver glycogen and blood sugar values than unirradiated, fasted animals. Although vaginal indications of estral activity persisted after irradiation, these elevations of carbohydrate levels could not be correlated with estral behavior. Presumably they are correlated with the radiation-induced increase in activity of the pituitary-adrenal system.

Figure 1. Irradiation of vertebral column. The rat was exposed to 3500 r of x-rays in the region indicated by the arrows in the radiogram. Six months later the animal was incontinent and unable to use the paralyzed hind legs.



### Effect of Irradiation on the Spinal Cord

Localized irradiation of the vertebral column of rats with 3500 r produces, with some consistency but only after an unpredictable latent period of 5 to 9 months, incontinence of urine and a motor weakness of the hind legs that develops inevitably into posterior paralysis. The initial lesion resolves into extensive destruction of white matter with little neuroglial reaction and almost intact gray matter in the region of the spinal cord related to the area of irradiation. It is doubtful that, as is commonly believed, the damage is secondary to damage of blood vessels and consequent lack of oxygen and nutrient. A comparable clinicopathological process has also been produced in two monkeys. The work is being continued with particular attention to early effects and extension of the work with monkeys, which will permit more detailed neurological and pathological study than is possible in the rat. This problem has considerable significance in human neurology and radiotherapeutics, since the hazard of exposure to the spine is well known, but not adequately understood.

### Effect of Radiation on Proliferative Cell Systems

An important class of radiation syndrome concerns cell systems that depend for integrity upon continuing proliferation, for example, the intestinal epithelium, bone marrow, and skin. Study of this class of syndrome involves three major questions: (1) how does radiation interfere with cell production; (2) how does the cell population respond to interference with cell production; and (3) what are the consequences to the whole organism of damage to cell systems? The effects of radiation on intestinal epithelium have been intensively studied within the framework of the first two questions and the areas of biology concerned, cellular and molecular biology, and population kinetics. Of particular moment is the finding of adaptation under repeated or continuous irradiation. The phenomenon is not attributable to selection of resistant cells but is linked to nucleic acid synthesis without cell division and a homeostatic mechanism which controls number but not quality of cells. Similar study of hair follicles has been made. Deoxyribonucleic acid (DNA) synthesis by the proliferative cells of the follicle, as found for those of the intestinal epithelium, decreases shortly after irradiation and is resumed, after high doses, prior

to or without return of mitotic activity. Production of DNA (measured by the incorporation of labeled thymidine) is reduced by about one-half within minutes after irradiation; further reduction occurs within hours after treatment. Since the doubling time for DNA normally is about six hours, the early decrease after irradiation is attributable to inhibition of synthesis rather than to inhibition of the entry of cells into the synthesizing stage.

#### **Hormonal Control of Glucose and Fatty Acid Metabolism**

The glucose present in the circulating blood of the fasting animal is in transport from the liver for use by the other tissues. A similar role has recently been envisaged for the nonesterified fatty acids of the blood, their source being the adipose tissue depots and their fate early combustion by other tissues. A great many hormonal factors influence both supply and demand for circulating glucose, and several factors are already known which influence the supply of circulating nonesterified fatty acids. These general problems are being studied, in collaboration with a group at the New York University College of Medicine, by use of  $C^{14}$ -tagged glucose and palmitic acid.

Conflicting evidence as to whether insulin directly inhibits glucose output by the liver led to experiments on dogs which clarified this situation. When glycogen breakdown is taking place in the liver, insulin does interfere with glucose output, whereas in the dog kept on a mixed diet (but not on a high carbohydrate diet) prior to fasting, this influence of insulin is scarcely seen, and the predominant immediate action of injected insulin is to increase glucose uptake by the tissues. The existence of the just-described effect of insulin on the liver has profound theoretical significance, for it cannot be explained on the basis of insulin's facilitating glucose transport across the cell membrane. The latter concept has been held during the last ten years or so to be an adequate description of the immediate effect of insulin on glucose metabolism.

With regard to nonesterified fatty acids, evidence has been accumulated indicating that the turnover of these acids in the blood is directly proportional to their concentration over a tenfold range. All points collected fall on the same straight line, even though nine hormonally modified or drug-treated states are encompassed by the observations. It thus seems possible to use nonesterified fatty acid concentration as a direct measure of the

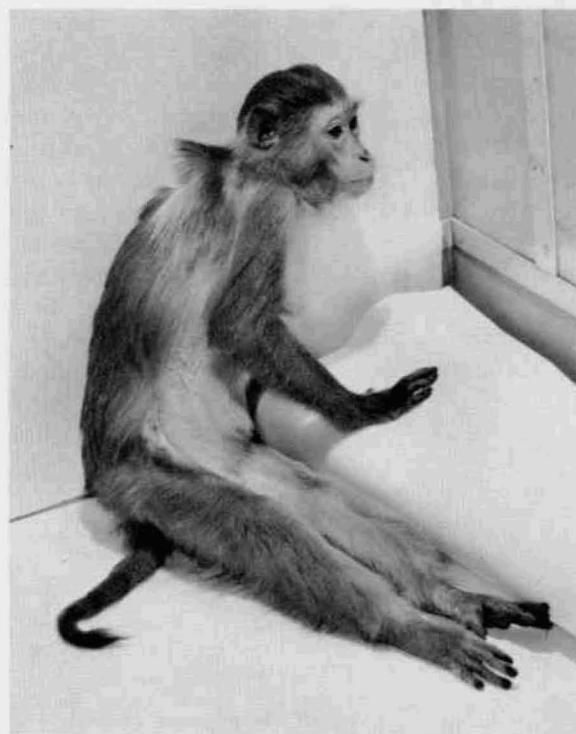
rate of turnover. Hormonal effects thus far noted are confined to changes in the rate of release of nonesterified fatty acid from adipose tissue and do not affect the rate of uptake except in a way ascribable to the changes in nonesterified fatty acid concentration in the blood.

#### **Antibody Specificity**

The solution of the problem of antibody specificity is important for an understanding of the immune response and its damage by radiation. The problem can be divided into two main areas for investigation. The first is the determination of the structure of the active site which allows a given antibody to react only with its own particular antigen. The second is the determination of the mechanism by which antibody is formed, whether by a recoiling of the chemically related component of blood, gamma globulin, or by a true genetic modification.

Promising results have been obtained in both areas. A general method consisting of three steps

Figure 2. Monkey irradiated with 3500 r of x-rays in the lower neck region. Six months later weakness became evident, followed by paralysis of the hind legs. Severe malacia of the irradiated spinal cord was found at autopsy.



has been developed to label the active site of antibody: (1) treatment of the antibody in the presence of hapten with a specific unlabeled reagent, (2) removal of the hapten, and (3) reaction of the groups at the active site with the same reagent but with a radioactive label incorporated. The method has been applied successfully to introduce radioactive iodine into the active site of antibodies directed against the *p*-azobenzenearsenate grouping.

When the amino acid content of the labeled antibody was compared with that of untreated antibody, it was found that only tyrosyl and histidyl groups had reacted with iodine under the conditions employed. A second protein reagent, photooxidation, which selectively destroys histidyl residues, was then used. It was found that antibody activity was not altered by the photooxidation of 11 of the 16 histidyl groups, but was lost on subsequent iodination when only tyrosyl residues were destroyed. Thus, tyrosine was identified as the iodine-reacting amino acid at the active site of the anti-arsanilic acid antibody.

Since the active site of anti-arsanilic acid antibody contains an amino acid that binds iodine, the iodination reaction was used to investigate the second aspect of antibody specificity, the relationship between antibody and gamma globulin. Purified preparations of these proteins were exposed to successive additions of iodine in both small and large amounts until saturation of the surface reactive groups was achieved. In all cases the uptake of iodine by antibody was so nearly identical to that of gamma globulin that any difference in the surface configuration must be limited to the area of the active site, i.e.,  $\approx 2\%$  of the total surface.

Furthermore, when the amino acid content of the comparably iodinated antibody and gamma globulin was assayed, the amino acid composition and the number and kinds of residues which had reacted with iodine were found to be identical in both, within the experimental error of the method. These experiments indicated, therefore, that antibody specificity must result from very small structural changes, within the order of two amino acids.

## GENETICS

### Genetic Control of Tobacco Tumors and Effects of Radiation

The genetic control of tumor formation in tobacco hybrids is indicated by the segregation observed for the presence and absence of tumors as

well as different expressions of the tumorous condition in hybrids between  $F_1$  *Nicotiana langsdorffii*  $\times$  *N. sandarae* and five other species combinations. Further confirmation of genetic control is afforded by linkages obtained between factors for small flowers and tumor formation and for large flowers and lack of tumors. A nontumorous mutant of *N. glauca langsdorffii*, induced by x-raying seeds, gave  $F_1$  hybrids with no tumors when crossed with the tumor-forming amphidiploid. The subsequent segregating generations confirmed the dominance of the mutant nontumorous condition. The genetic results as a whole support the interpretation that tumor formation in tobacco hybrids is controlled by hereditary factors which show typical properties of segregation, linkage, and mutation.

A number of different genotypes involving the species *N. glauca* and *N. suaveolens* inbred with genomes or gene recombinations of *N. langsdorffii* and *N. sandarae* were grown for  $\approx 108$  days under 5 to 400 r of gamma radiation per 20-hr day. Certain genotypes, including the radiation-induced, nontumorous mutant, produced no tumors at any level of irradiation, including the highest (which approached lethality), or when not irradiated. Others produced tumors at all dosages, and also when not irradiated. A third group formed tumors late in the season only when irradiated at levels of 200 to 350 r per 20-hr day, depending upon the genotype.

### Induced Variation in Symbiotic Nitrogen-Fixing Bacteria

Perhaps the most important natural conversion of atmospheric nitrogen into a form available to higher plants is carried out symbiotically by bacteria living within nodules on the roots of host plants. Effective nodulation on alfalfa, peas, beans, and clover defines the four species of *Rhizobium* used in this work, respectively, *R. meliloti*, *leguminosarum*, *phaseoli*, and *trifolii*. Nodulation by *R. meliloti* on the hosts defining the other species is relatively rare, as is the converse, but the other more closely related species occasionally cross-nodulate on nonspecific hosts, except alfalfa. The immediate objective of the present work is to determine the feasibility of altering the host range and nitrogen-fixing efficiency of the bacteria by the techniques of experimental genetics. A more general objective is to gain information concerning the evolutionary relationship of plant-pathogenic and symbiotic nitrogen-fixing bacteria. The pro-



cedure involves marking cultures of related species of rhizobia with readily identifiable characters, e.g., antibiotic and phage resistance, and attempts to induce interspecies cross-nodulation via mutation, transduction, transformation, or recombination.

The following summarizes the work carried out thus far. The relation between survival and dose of ultraviolet (UV) and x-rays is nonlinear for log-phase cultures of the cultures tested. The  $LD_{50}$  ranges from 1.5 to 3.5 kr for x-rays and from 150 to 1400 ergs/mm<sup>2</sup> for UV. A number of antibiotics and antibacterial drugs, in disc tests, distinguish between some cultures or strains and thus provide

a degree of differentiation suitable for secondary markers. Differentiation between the species *R. leguminosarum*, *phaseoli*, and *trifolii* was not consistent, but several antibiotics separate this group of species from *R. meliloti*. Resistance to dihydrostreptomycin and erythromycin suitable for primary marker purposes was obtained in both control and irradiated cell populations. The expression of induced mutation followed a phenotypic lag. The frequency of induced resistance to dihydrostreptomycin exceeded the spontaneous level by  $\approx 15$ -fold with UV and 4-fold with x-rays at 90% lethality. The resistant mutants showed no evidence of antibiotic dependence. Bacteriophage

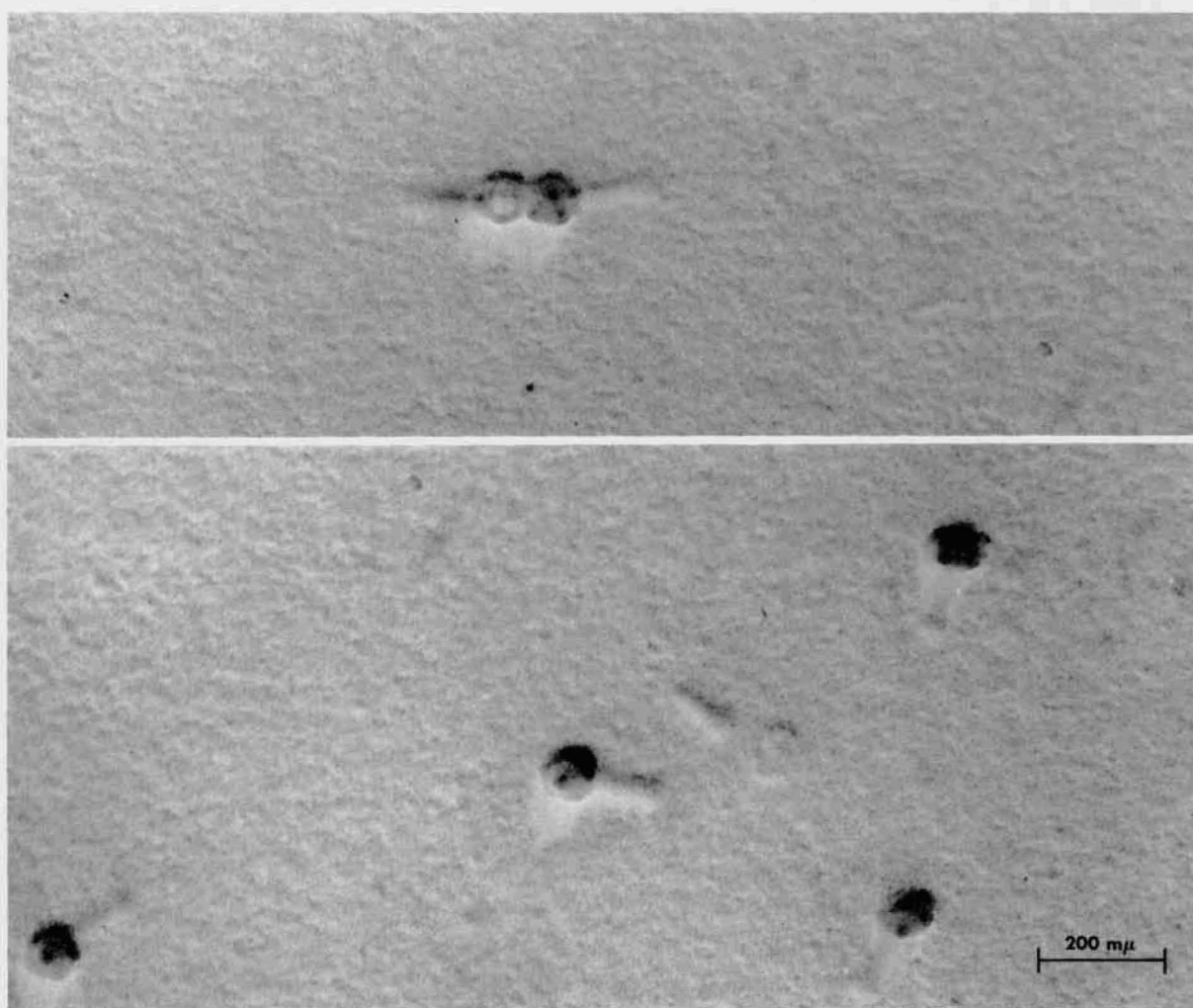


Figure 3. Electron micrographs (71,600 $\times$ ) of particles of rhizobiophage strain 6A, obtained from lysate of a *Rhizobium trifolii* culture. The preparation was shadowed with platinum-palladium and coated with a vaporized carbon film. The nature of apparent inclusions in the heads of the particles, seen as dark, round objects at the periphery of the head membrane, has not been determined.

isolated from field collections of nodules differentiated *R. meliloti* from the group comprising the other three species, but did not separate cultures within the group. The 90% absorption time and latent period for two phage strains were  $\approx 50$  and 80 min, respectively; the burst size was  $\approx 145$ . One lysogenic culture of *R. leguminosarum* has been found thus far. The search for lysogenic strains was complicated by a fairly high incidence of intercultural antagonism; tests have been devised that distinguish between such antagonism and the lytic effect of phage. The nature of the antagonism, which resembles the behavior of weak or unstable bacteriocins or antibiotics, is being investigated.

Nodules have been recovered from pea plants inoculated with x-irradiated cells of a streptomycin-resistant strain of *R. trifolii* that normally nodulates on clover but not on peas. The apparent mutants from the nodules retained the resistance marker and upon reinoculation formed nodules on both peas and clover. The nodules on peas did not fix nitrogen effectively, and those on clover were less effective than controls. The change induced by the irradiation appears to involve gain in host range with concomitant loss of nitrogen-fixing ability. The loss of effectiveness is consistent with other reports concerning the instability of this character.

#### **The Lysogenic Interaction Between Bacteriophage and Bacterium**

Bacteria infected with a temperate bacterial virus or bacteriophage may be destroyed, progeny phage being liberated in the process. Or they may survive and give rise to a hereditarily changed bacterial clone immune to subsequent infection, but able to produce 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 genetic control of the hereditary changes. The altered state of the bacterium is known as lysogeny, and prophage designates the controlling entity of this condition. Much was learned earlier about the genetic control of the ability to become prophage. At present, attention is being directed to the question of the maintenance of the lysogenic condition. Certain evidence suggests that the presence of prophage results in the formation of a specific immunity substance or repressor which represses production of virus by newly infecting bacteriophage and by the

prophage itself. The antibiotic chloramphenicol, which inhibits bacterial protein synthesis, enhances the lysogenic condition, which suggests that the repressor is not a protein. On the other hand, the antibiotic mitomycin C, which selectively inhibits bacterial synthesis of deoxyribonucleic acid, and the analogue of thymidine, 5-fluorodeoxyuridine, cause induction of phage production. Thus, the repressor may be a nucleic acid.

#### **Gene Structure and Mutation**

The chromosome in higher organisms is considered to be a multistranded complex of helically oriented molecules of the heredity determinant, deoxyribonucleic acid (DNA). This suggests the hypothesis that the gene also is a multigenic complex. Each gene on a single chromosome is visualized as a number (8, 16, 32) of identical multigenes, each occupying a comparable position on a DNA helix or strand within the chromosome bundle. Change at the strand level results in *multigene mutations* (e.g., 16 A mutates to 15 A + 1 A'). The multigene mutations do not gain expression except as the mutant strand increases in frequency within the bundle and ultimately monopolizes the multistrand complex (15 A + 1 A'  $\rightarrow$  ...  $\rightarrow$  16 A').

The hypothesis promises increased understanding of certain genetic enigmas. For example, the paucity of "point" mutations may result from change at the multigene level and subsequent slow release. The variability in doubled haploids may be due to summation and release of hidden multigenic variability. The hypothesis suggests further caution in the evaluation of the genetic hazards of radiation. Classic radiation-induced mutations arise from simultaneous damage (primarily breakage?) to all or most strands and require much more energy than is needed for change at the multigene or single-strand level. It would be expected that radiation-induced damage at the multigene level, like the changes leading to "point" mutations, would be slowly expressed as mutations in contrast to the relatively prompt release of classic mutations.

#### **Cooperative Radiation Mutation Program**

The goal of the Cooperative Radiation Mutation Program is to assess the usefulness of radiation in producing mutations that represent crop improvements. Whole plants, seeds, and cuttings are irradiated with x-rays,  $\gamma$ -rays, or neutrons at

Brookhaven and are then studied by cooperating groups in this and other countries.

One of the most interesting findings reported this year was that of Miss Charlotte Pratt of the New York State Agricultural Experiment Station in Geneva. Many commercial fruit trees are cytochimeras, that is, they possess different numbers of chromosomes in different cell layers of the stem apex. Thus, in a 2-2-4-4 apple chimera, cells of the two outer layers of the apex have the diploid chromosome number, but the inner layers have cells with a tetraploid chromosome complement. Such a plant will produce normal haploid ( $n$ ) gametes. One of the objectives of apple breeding research is to produce triploid and tetraploid plants which may have beneficial new characters by virtue of the additional sets of chromosomes. For this purpose diploid rather than haploid gametes are required.

Following x-irradiation of apple scions and grafting onto unirradiated stocks, cytological examination of the growing shoots revealed that the usual 2-2-4-4 chimera had been disrupted and a variety of new chimeral types had been produced. Among these were several with a 2-4-4-4 constitution, which are expected to yield the diploid gametes required by the breeding program. In other instances 2-2-2-4 and 2-2-2-2 types were found. It is believed that a differential radiation sensitivity, based upon both the chromosome number and the location of the different ploidy levels, has resulted in selective cell destruction followed by regeneration of a new stem apex from the surviving cells.

## CELL PHYSIOLOGY

### Alteration of Radiation-Induced Chromosome Damage

The damage to chromosomes produced by irradiation of cells depends to an appreciable extent upon the environment during and after the irradiation. The physiological and biochemical basis of such dependence has considerable bearing on attempts to understand and modify radiation effects in whole organisms. The following results suggest an explanation for the effect of certain environmental conditions. *Habrobracon* oocytes exhibit less damage when irradiated under nitrogen or carbon monoxide than under room air, an expression of the well-known enhancement of the effect of irradiation by oxygen. However, exposure to nitrogen

or carbon monoxide after irradiation in air increases the degree of damage. This suggests that some repair mechanism requiring oxygen is operative after irradiation. The presence of a gas mixture containing carbon dioxide and oxygen during irradiation increases chromosome aberrations in *Tradescantia* microspores. The effect of the gas mixture endures for  $\approx 20$  min after its removal if it is replaced by room air, but not if it is replaced by nitrogen or a vacuum. There is reason to believe that the carbon dioxide treatment decreases the viscosity of protoplasm, which would facilitate movement of chromosomes and, thus, production of aberrations. However, the requirement for oxygen suggests that energy is required for the movement of chromosomes. Again in *Tradescantia*, maintenance of irradiated microspores in the cold for prolonged periods results in a marked decrease in chromosome aberrations. Presumably, the movement of chromosomes is decreased in the cold, and fewer exchanges between broken chromosomes occur.

### Relationship Between Chromosomal, Nuclear, or Cellular Size and the Radiosensitivity of Different Plant Taxa

The causes of the very great differences in the amount of ionizing radiation that can be tolerated by different species has long been a central problem in radiobiology. Despite many investigations, it has not been possible to explain these differences or to predict the tolerance of a species or form not previously studied. However, the results of a ten-year study of various species of higher plants grown under chronic irradiation in the Brookhaven gamma radiation field have provided information that appears to allow fair prediction of the tolerance of a given plant to ionizing radiation.

More than 200 species representing some 50 plant families have been studied. The plants were exposed acutely or chronically to gamma radiation from radiocobalt. Plants of each species studied were exposed simultaneously at different distances from the source. In this way a wide range of dose rates (or dosages) were given, and after one or two test runs it was usually possible to determine the dosage necessary to stunt severely or stop the growth of the plant. In the species studied, it was found that tolerances differed by several hundredfold. For instance, pitch pine (*Pinus rigida*) is killed by prolonged exposure (6 to 8 yr) at dose rates as low as 5 r per day, whereas common *Gladi-*

*olus* can grow despite exposure to 5000 r per day for several months. Some of the most resistant plants were polyploids (having more than two sets of chromosomes). When only diploids (plants with two sets of chromosomes) are considered, the differences in sensitivity were still at least 250-fold.

Many types of measurements were made. Somatic mutations were scored in petals of plants

severe growth inhibition, or even death. There is a correlation between the acute and daily chronic doses necessary to produce severe growth inhibition in young plants of different species. (The acute dose averages  $\approx 13$  times the daily chronic dose.) Two polyploid species (*Chrysanthemum* and *Sedum*) show a protective effect proportional to the degree of polyploidy. From this study it appears



Figure 4. Facility for irradiation of plants under controlled environmental conditions. The chamber in the background provides for growth under prescribed conditions of illumination, temperature, and humidity. A radioactive cobalt source is housed within the lead shield in the foreground.

heterozygous for flower color (such as snapdragon or petunia). The results indicated that the color change resulted from chromosome breakage. The yield of mutations was reduced by dose fractionation. The frequency per roentgen of somatic mutations (and of chromosome deletions) is higher during periods of slower growth than periods of fast growth. Evidence of long-term cumulative dosage effects were found in *Pinus* (pines) and *Taxus* (yews). Over a long period (several years) at dose rates as low as 5 r per day, cumulative effects are expressed as severe morphological deformity,

that in diploid plants the sensitivity to chronic radiation is directly related to the average nuclear volume of the cells of the apical meristem – the larger the volume, the greater the sensitivity. It also appears that, for a given volume, increase in chromosome number without polyploidy decreases sensitivity. The effect of such increase in chromosomes is greater than that obtained with ordinary polyploidy.

Analysis of the relationship between sensitivity of different species and nuclear volume indicates that a constant (or nearly constant) number of



ionizations per nucleus is required to produce growth inhibition or death. This relationship supports the concept that a cluster effect such as chromosome breakage (deletion) may be the critical event or events. Differential sensitivity results in part from the different doses required to produce a unit number of breaks in nuclei of different average volumes. The protective effect associated with increasing chromosome number or polyploidy is assumed to result from the reduced seriousness of an average break or deletion as the chromosome number increases.

Clarification of the manner in which radiosensitivity is determined in different species should make predictions of radiosensitivities possible in the very near future. This may hold true for animal species as well as for plants. Extrapolation of certain kinds of radiobiological data from one species to another can now be made on a reasonably sound basis.

#### The Submicroscopic Basis of Photosynthesis

Photosynthesis, the biological process that uses solar energy for synthesis of organic matter from carbon dioxide and water, involves the capture of light energy by the photosynthetic pigments, the conversion of light into chemical energy, and, finally, use of the newly generated chemical energy for synthesis of cell constituents. In higher plants the several events occur within complex organelles, the chloroplasts. For analysis of the light-dependent reactions a simpler system would be preferable. From this point of view the photosynthetic bacteria, particularly those dependent upon light as the external source of energy, are of special interest.

In the purple sulfur bacterium, *Chromatium*, the capture of light and conversion of its energy to chemical energy was found earlier to occur within the chromatophore, the simplest and smallest organelle known to participate in photosynthesis. Recent work with the green sulfur bacterium, *Chlorobium thiosulfatophilum*, also an obligate phototroph and anaerobe, indicates that photosynthesis can proceed at a level of organization even lower than that of the *Chromatium* chromatophore. *Chlorobium* resembles nonphotosynthetic bacteria such as *E. coli* in the number of subcellular components which can be identified in the analytical ultracentrifuge and in structures observed in electron micrographs of thin sections of the organism. Three major components with sedimentation coef-

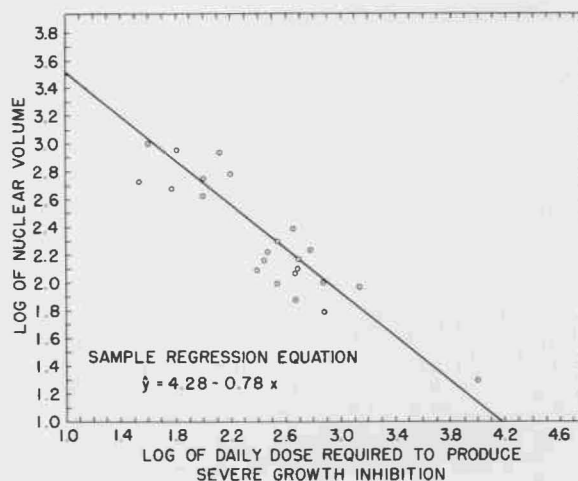


Figure 5. The relationship between nuclear volume and radiosensitivity in 24 species of plants.

ficients of  $\approx 5$ , 30, and 50 S have been obtained from extracts of the bacterium. The 50 S component bears the photosynthetic pigments and can be further purified by centrifugation. As a first approximation these purified particles have a molecular weight of about one million and contain  $\approx 50$  molecules of chlorophyll; thus, these particles are about an order of magnitude smaller than the *Chromatium* chromatophore. Although more work is required to characterize the *Chlorobium* particle, it may well approach the lower limit of organization essential for the initial photosynthetic event.

#### Ribonucleic Acid Metabolism in Dividing Cells

Ribonucleic acid (RNA), a major component of cells, is present in nearly all structures. It is especially concentrated in the nucleolus, a dense body in the nucleus, and occurs in the cytoplasm as a component of certain particles, the microsomes, and probably also in solution. It appears to be involved in such basic functions as protein synthesis, differentiation, and development. RNA is chemically related to deoxyribonucleic acid (DNA), which is located in the chromosomes and is concerned with heredity, the process by which specific patterns of cellular activity are held constant and passed to progeny. Thus, DNA appears to serve passively as a master template that stores genetic information, while RNA serves as the intermediate which transfers information to sites of protein synthesis.

The metabolism of RNA by cells of the growing root tip of *Vicia faba* has been studied by allowing

cell division to take place in the presence of tritium-labeled cytidine, a precursor of both nucleic acids, and then determining the localization of the precursor within the cell by autoradiography. The results obtained indicate that during cell division RNA is synthesized in both the chromosome and nucleolar portions of the nucleus, the

RNA produced in the chromatin moves to the nucleolus, and then both chromatin and nucleolar RNA move to the cytoplasm. The findings support the impression that RNA serves as a carrier of genetic specificity from the DNA of the chromosomal genes to sites of protein synthesis in the cytoplasm.

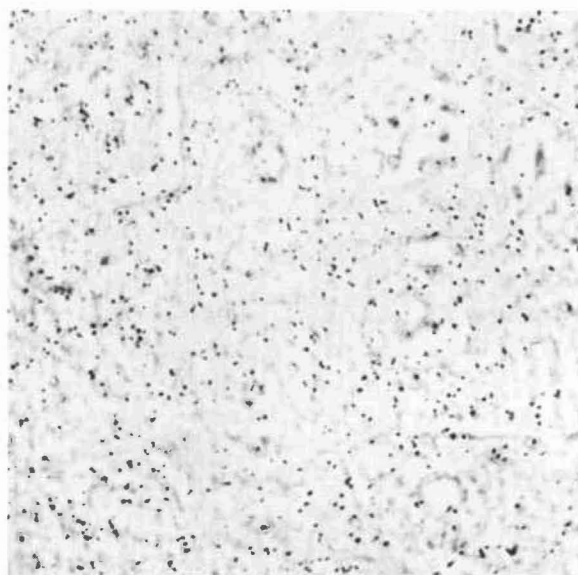
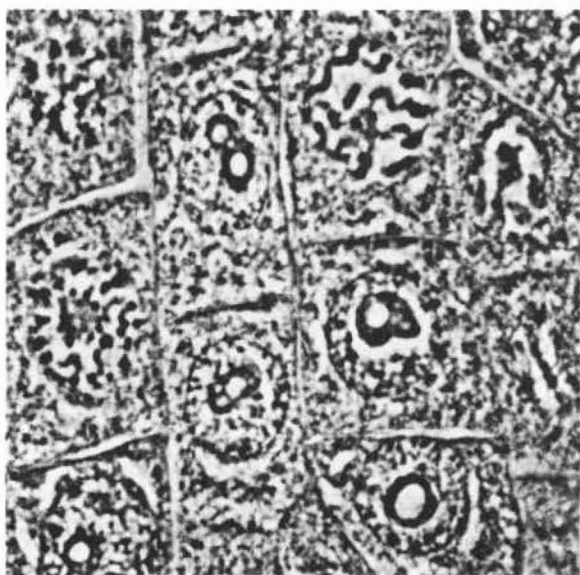
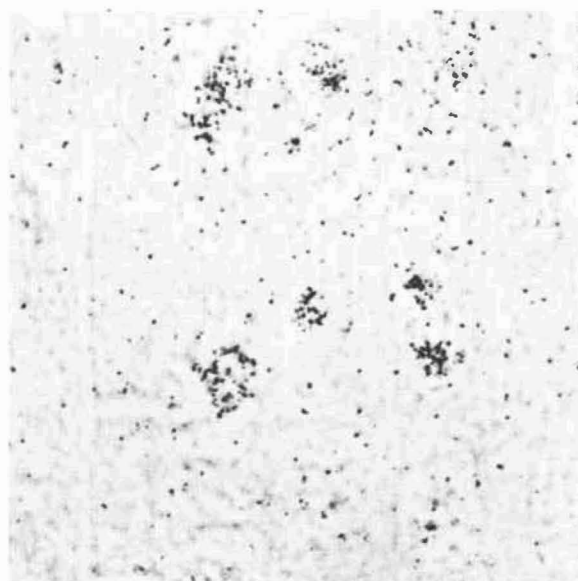
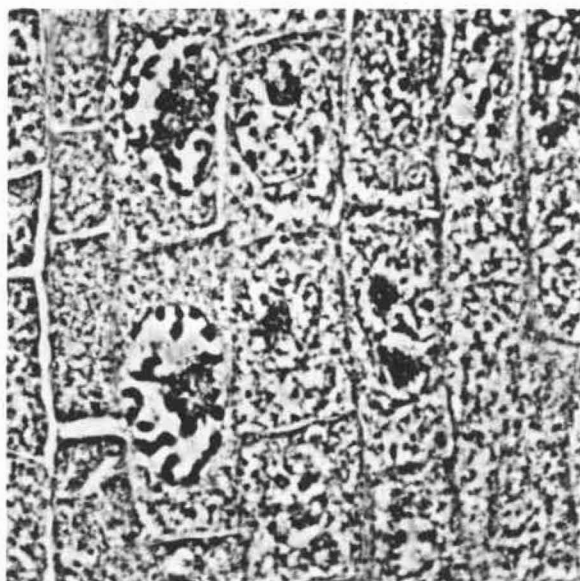


Figure 6. Migration of RNA is shown in these photomicrographs of root cells from a bean plant. Cells at top were grown in a medium containing a labeled precursor of RNA. Radioautograph (top right) shows RNA concentrated in the nuclei of the cells, specifically in the nucleoli. After cells are transferred to unlabeled medium (bottom), uniform distribution of labeled RNA shows that it has moved into the cytoplasm from the nucleus.

### Pollen Grain Germination

Pollen grains of flowering plants are of two types, one having two nuclei, the other three. The binucleate grains will germinate *in vitro* while the trinucleate ordinarily do not. The germination of binucleate grains *in vitro* proceeds readily if concentrated populations are sown, but decreases as sample concentration decreases. This dependence of germination on concentration appears to be a reflection of a dialyzable, relatively heat-stable factor obtained from pollen that triggers germination. The nature of this material is under examination. Inference and early tests suggest that trinucleate pollen grains lack a germination-triggering factor, but that such a factor is present in the stigma of the plant.

### Relation Between Porphyrin and Heme Synthesis

Duck erythrocytes *in vitro* synthesize protoporphyrin. Of the total porphyrin produced, a small fraction appears in a free form, the rest as the iron complex, protoheme. Both forms of the pigment derive from the same precursors. Thus, the simplest hypothesis explaining the relation between the two forms would be that free pigment formation represents that part of the total not coupled by the cell to iron. However, certain findings appear to require a more elaborate hypothesis. Among the findings are the following: As the erythrocyte ages it loses the ability to take up iron and thus makes less heme, but free porphyrin production changes little, if at all. Increase and decrease in free porphyrin production without marked change in heme synthesis result from the presence of particular substances of biological interest. Although these findings suggest that the hypothesis indicated is not sufficient to describe the relationship between porphyrin and heme synthesis, they as yet do not furnish an elaboration.

### Inactivation of Bacteriophage by Ultraviolet Light

Bacteriophage suspended in water is more sensitive to ultraviolet light when irradiated in the frozen state ( $-20^{\circ}\text{C}$  and lower) than when irradiated at room temperature. The increased sensitivity does not occur with dry phage or phage suspended in glycerol. The effect is obtained with 2537-A radiation, but not with 2894-A, which implicates nucleic acid but not protein in the effect. The possibility that a photochemical alteration of

the nucleic acid may occur at low temperatures is being explored.

### Auxin Studies

Studies were continued during the year on the endogenous hormones (auxins) of the Lombardy poplar bark and their relationship to the previously described phenomenon of seasonal polarization of root emergence. Roots are present in the bark of the Lombardy poplar as a normal developmental feature. Cuttings taken from trees during the period of dormancy, from October through April, and placed in darkness, will produce roots uniformly along their entire length. Cuttings made during the period from May through September will produce roots only at their basal end, despite the fact that root primordia are known to be present along the length of the cutting. With the aid of Dr. and Mrs. J.P. Nitsch of the Laboratoire du Phytotron in Gif-sur-Yvette, France, the auxins were extracted from polarized and nonpolarized cuttings, separated by paper chromatography, and assayed by using the oat second-internode test developed by the Nitsches. No differences, either qualitative or quantitative, could be detected. However, the application of exogenous auxins is capable of completely depolarizing polarized cuttings, which indicates that internal differences in auxin concentration or distribution actually do exist in the bark tissues. A new bio-assay procedure was developed in which the ability to depolarize cuttings was used as the criterion of effectiveness of extracted auxins. It was found that polarized cuttings contain enough extractable auxins completely to depolarize other polarized cuttings, which indicates either that the polarization is due to a differential distribution of the auxins so that they are not available to the root primordia, or that the auxins in dormant cuttings are chemically bound in such a form as to render them ineffective. The possibility also exists that the extraction procedure is producing auxins from their precursors. Future research will attempt to explore this further.

## BIOCHEMISTRY

### Structure of Ribonuclease

Ribonuclease is the first enzyme for which it is possible to write a formula showing all the covalent bonds in the molecule. The molecule can be pictured as an ordered and characteristic linear sequence of 124 amino acid residues. The poly-



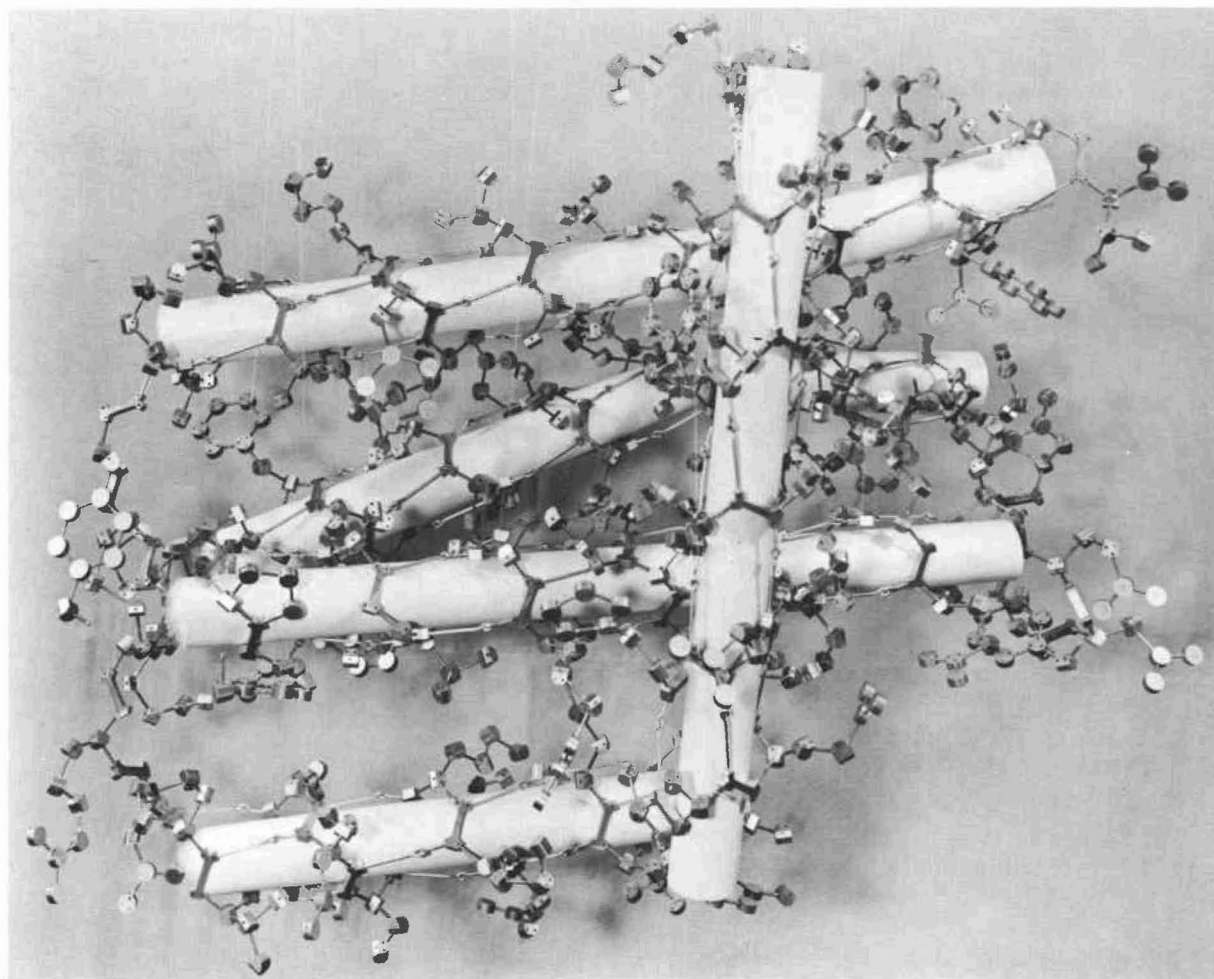


Figure 7. Possible secondary and tertiary structure of ribonuclease. Paper cylinders have been inserted into the helical backbone of the molecules.

peptide chain, through poorly specified interactions between various portions of the chain, is elaborately folded upon itself into a three-dimensional cluster. The spatial arrangement (tertiary structure) of the convoluted backbone and projecting side chains determines the ability of the molecule to bind its substrate in the specific orientation required to bring about chemical interaction at the catalytic center of the enzyme. To gain further understanding of the tertiary structure, the accessibility of the amino acid side chains to various chemical probes is being studied; "exposed" chains are probably more accessible to attack than "buried" ones. Alkylation with 2,4-dinitrofluorobenzene has proved of value in studying the lysine residues. Two of the ten lysines present are partic-

ularly sensitive to modification by alkylation. The alkylation of one of these is of special interest because it leads to the formation of an enzymatically inert, homogeneous product. Further study of this product should prove of value in delineating those portions of the molecule most intimately connected with the catalytic functions of the enzyme.

#### Muscular Contraction

The contractile protein, actomyosin, and the energy source, adenosine triphosphate (ATP), are involved in muscular contraction, but the manner in which ATP transfers its energy to the protein has eluded detection. Evidence has been obtained by using  $O^{18}$ -labeled water that the hydrolysis of ATP by actomyosin involves a phosphorylated

protein intermediate. Further study to characterize this intermediate has been carried out. The metal ion specificity of the  $O^{18}$  exchange reaction was found to be roughly dependent on the radius of the metal ion. A maximum was observed for the transition element  $Mn^{++}$ , with lower values for metals of smaller and larger radii. An interesting factor in this dependence was the correlation with metal ion specificity for the contraction process. This tends to support identification of the intermediate as a step in this contraction.

Treatment of the myosin protein with trypsin results in a new protein, heavy meromyosin, which is able to hydrolyze ATP but not able to contract. This protein has roughly the same metal ion specificity as the intact myosin. The minor deviations indicate that the amino acids deleted by the action of trypsin have some influence on the active site. The results suggest a site for ATP hydrolysis which provides energy for contraction, but hydrolysis can occur even when the protein has been damaged to such an extent that the energy can no longer be used for contractions.

#### **Photosynthetic and Respiratory Enzymes in Leaves**

The development of enzyme activity is of considerable significance for understanding the metabolic organization of cells. To define the involvement of the many enzymes of green leaves in photosynthesis, in respiration, or in both, the activities were studied as a function of stage of growth and photosynthetic activity. One group of enzymes [isocitric malic, glucose-6-phosphate, 6-phosphogluconic, and lactic dehydrogenases, glutathione and glyoxylic reductases, phosphoenolpyruvate carboxylase, aconitase, enolase, phosphofructokinase, and malic enzyme] does not appear to function in photosynthesis. The activities of these enzymes reached peaks earlier than known photosynthetic activities and, at the developmental stage when maximum photosynthesis occurs, were less than required for participation. A second group [ribulose diphosphate carboxylase, triphosphopyridine nucleotide (TPN)-linked glyceraldehyde-3-phosphate dehydrogenase, and photosynthetic TPN reductase] is involved only in photosynthesis and presumably is localized in chloroplasts. A third group [phosphoglyceric kinase, DPN-linked glyceraldehyde-3-phosphate dehydrogenase, triosephosphate isomerase, aldolase, and phosphoglucose isomerase] appears to participate in both photosynthesis and respiration.

#### **Enzyme Structure and Function**

Radiation produces chemical modification of the side chains of enzymes and other proteins. To understand its effect, the role of these side chains in enzyme action must be evaluated. Such study is complex because of the large number of side chains in even the smallest enzyme. Recently, a new technique has been developed for the kinetic analysis of amino acid residue modification and loss of enzyme activity. This technique has been applied to two enzymes, phosphoglucose mutase and chymotrypsin, which have been the center of study for some years. Irradiation with visible light in the presence of methylene blue and oxygen was found to cause oxidations of the histidyl, methionyl, cysteinyl, tryptophanyl, and tyrosyl side chains of these enzymes. No other amino acids were affected. Kinetic analysis revealed that a single histidyl and a single methionyl residue were responsible for loss of enzyme activity in the case of each enzyme. Destruction of other residues had no effect on enzyme activity. Modification of the histidyl residue reduced activity by a factor of  $\approx 12$ , whereas oxidation of the methionyl residue reduced activity by a factor of  $> 200$ . Both residues appeared in two groups of unequal reactivities, which indicated that some were in protected positions in the interior of the molecule, whereas others were on its surface. The residues responsible for enzyme activity were on the surface. These studies not only give information about the activity of these two important enzymes, but also indicate that the kinetic procedure may be a valuable tool for interpretation of radiation and other damage to enzymes.

### **BIOPHYSICS**

#### **The Deuteron Microbeam as a Tool for Biological Research**

The 22.5-Mev deuteron microbeam, 25 to 250  $\mu$  in diameter, from the 60-in. cyclotron appears to be a promising tool for biological research, particularly in plant morphogenesis and allied fields. The beam has been used to hit specific targets in the embryo of dry maize seed. Marked stimulation in growth of the primary root was obtained when the stem apex region, extending from the base to the top of the first leaf, was irradiated. This effect was less evident, or probably insignificant, when the root itself was irradiated. With the beam used as a slit, lines "drawn" inside the embryo can be ob-



Figure 8. Maize seedling showing lines "drawn" on leaves by irradiation of the dry seed embryo with a 25- $\mu$  deuteron beam at points 200  $\mu$  apart.

served as yellow marks in the mature green leaf. In one experiment the embryo was irradiated with a 25- $\mu$  beam at four separate points 200  $\mu$  apart. The marks produced by the irradiation were clearly visible in the mature plant on the coleoptile and first three leaves. Comparison of the initial distance of 200  $\mu$  between the marks and the distance between the marks in the mature plant indicated an evenly distributed, 15-fold increase in length of the coleoptile during the transition from embryo to maturity. The first, second, and third leaves increased 42, 140, and 244-fold in length, respectively. The growth increment was much greater in the basal part of these leaves than in the distal half.

#### Biological Effects of Cosmic Rays

The biological effects of a beam of deuterons 25  $\mu$  in diameter reasonably simulate those of the secondary particles from a primary cosmic-ray "thin-down." The dose required to damage mouse brain when the microbeam is applied is about two orders of magnitude greater than when a beam of 1

mm or larger diameter is used. The most logical interpretation of this difference in dose requirement is that nerve cells are relatively resistant to radiation, while the capillaries are relatively sensitive. Thus, with a wide beam capillaries are destroyed, which indirectly leads to destruction of nerve cells. With the narrow beam the likelihood of hitting a capillary is much smaller, and it is possible to study the direct effect of radiation on the nerve cells. Such study indicates that cells die in a manner that differs from the usual pattern associated with capillary destruction. The cytoplasm disintegrates first, followed by breakup of the cell, rather than the reverse. This suggests that the radiation releases something inside the cell that leads to its destruction. Cytochemical procedures indicate the presence in the irradiated cell of a proteolytic enzyme, possibly liberated from lysosomes abundant in neural cytoplasm.

#### Membrane Permeability

Membrane permeability is a basic biological problem that has been under study for many years. Tests of certain older theories of permeability made possible by the use of isotopic tracers have given results at variance with prediction. This has raised questions not only about the theories but also about the interpretation of some kinds of isotope experiments. Both aspects of the problem continue under theoretical investigation. Criteria for evaluation of data from isotope experiments have been developed, and it has been found that the description of the transfer process through membranes attained by the methods and concepts of statistical mechanics can be derived more easily by application of Newtonian mechanics. Experimental testing by the theoretical relations is in progress.

#### Inactivation of Enzymes by Radiation

Of the various physicochemical changes produced in enzymes and other proteins by radiation, those leading to loss of biological activity are of particular interest. Postulated modifications associated with inactivation of enzymes may be grouped into three categories: (1) destruction of an amino acid residue required for activity, (2) *generalized* disruption of bonds responsible for enzyme conjunction, and (3) *specific* as opposed to indiscriminate changes in conformation. Considerable evidence has been obtained to support a "weak link" hypothesis, one of the possibilities in-

cluded in category (3), which proposes disruption of specific, easily broken bonds that as a group are critical for biologically active conformation. It is considered that most of the bonds involved can be ruptured singly by the energy associated with an ionizing event or ultraviolet quantum. Since such energy is of thermal magnitude, the thermal content of the protein molecules can contribute to the disruptive processes. The cluster of bonds making up the "weak link" are presumed to be disulfide bonds and a limited number of other intramolecular bonds. Disulfide bonds are proposed because they are essential to the activity of many proteins and are ruptured by small amounts of energy. Rupture of all bonds of the group is thought to produce irreversible inactivation, whereas incomplete rupture constitutes reversible inactivation.

Current results indicate three classes of trypsin molecules in solution after irradiation: active, damaged, and inactive. Operationally defined, *active* molecules show activity when exposed to urea or heat treatment before addition of substrate for assay; *damaged* molecules show activity if ex-

posed to substrate first after irradiation, but are *inactive* if urea or thermal treatment precedes addition of substrate. Since the quantum yield for inactivation of trypsin with ultraviolet (2537 Å) is roughly one molecule per 100 quanta, the active molecules may have sustained some radiation damage. However, in terms of the above hypothesis, the weak link in active molecules has been essentially unaffected, while the damaged molecules are thought to exhibit partial disruption of the weak link and to be, thereby, in a reversibly inactivated state. Damaged molecules appear to contain one to three more hydrogen bonds than those of the inactive class and bear one disrupted cystine instead of two. As indicated by the operational definition, damaged molecules show activity if treated with substrate before assay, but if exposed to *p*-chloromercuribenzoate before substrate, reactivation is prevented. This, in accordance with hypothesis, implies that disruption of a cystine group and the associated conformational changes represent an essential step in the inactivation of trypsin by irradiation.

# Medical Research

Fiscal 1960 saw a new height in the Medical Department's activity since its organization in 1949. Collaboration between members of the scientific staff was extensive, in spite of the fact that the program ranged from basic activities to applied research and included technological development. A group of senior investigators has developed, skilled in the art of medicine and increasingly versed in nuclear physics as it applies to that art.

The present report is concerned with work completed or very clearly defined within the year. It constitutes only a sketch of the Department's activities, but an attempt has been made to select illustrative trends.

## CANCER AND ALLIED DISEASES

As previously emphasized, the program of the Medical Department is not primarily concerned with research on cancer, in spite of the intimate relationship of cancer to many of the researches. The use of radioactive isotopes in the treatment of malignant disease has provided an opportunity to study the control and movement of radioactive isotopes within the body. In turn, this knowledge is important not only in the proper utilization of radioactive isotopes for therapy but also in the evolution of diagnosis and in the elucidation of basic life processes.

## REACTOR RADIOLOGY

### Neutron Capture Therapy

The largest single effort in the Department is the development of neutron capture therapy for control of malignant intracranial neoplasms and study of the effects upon cellular structures of neutron infiltration.

**Medical Research Reactor.** The Medical Research Reactor (MRR) was subjected to an extensive investigation to define its emergent neutron cloud spectra at a point in free air. Fifteen different geometries were used. The cadmium ratio was employed wherever applicable. In addition, gold, cobalt, copper, enriched  $U^{235}$ , depleted  $U^{238}$ ,

aluminum, and sulfur radioactivants were used. In testing the radiological characteristics of the radioactivants, collaborative arrangements were made with Oak Ridge National Laboratory. The effort has been extensive, and it is hoped that it will result in characterization of the neutron cloud and development of a suitable and simplified procedure for further testing of this type.

The MRR was tested at various power levels up to and including 5 Mw. The neutron output appeared to correlate well with the power levels attained. A semipulsed type of operation was tested for possible use as a means of shortening patient exposures. The reactor moderating and reflector components as well as the gamma shielding were carefully studied in terms of the induced gamma radiation. A dozen different configurations were made available for study by altering the moderating and filtering materials in the shutter and reflector of the reactor. It is estimated that these studies will require at least two more years for completion. Furthermore, the problem of shielding must be fully investigated, since  $Li^6$  metal seems to be the only useful material found to date for limiting the neutron field.

This effort has led to scrutiny of the information upon which physical dosimetry is based. The problem has become increasingly complex as better information was sought. Collaboration is under way with the Applied Mathematics Division to evolve suitable formulation of the problem for computer solution.

**Animal Experimentation.** Upon exposure of the rabbit's head to  $10^{13}$  thermal neutrons/cm<sup>2</sup> with a concomitant dose of  $<100$  r, no abnormal behavior of the animal can be seen initially. Several thousand mice were exposed to an intense thermal neutron dose, with and without accompanying infusion of  $B^{10}$  compounds. Exposure was limited to one hind leg, and no observable difficulty has thus far ensued.

In collaboration with the New York Eye and Ear Infirmary, the electrophysiological effects of thermal neutron exposure on the eye are being studied. The deleterious effects seen after exposure to x-ray did not occur with thermal neutron exposures. Special histochemical techniques are also



being used in order to detect any unexpected sensitivity of tissues to thermal neutrons.

Control of transplantable mouse tumors by the neutron capture procedure has been found fruitful. Emphasis was given to aspects of standardization of the reactor itself and to evaluation of neutron effects on the tumor. Indications are that animals may be cured of transplantable tumors if they receive sodium pentaborate and a thermal neutron exposure of  $3.2 \times 10^{12}$  neutrons/cm<sup>2</sup>-sec. Test animals have been observed for long periods of time and appear to be doing well. The spontaneous osteogenic sarcoma of a dog referred for treatment was greatly decreased by neutron capture therapy.

**Clinical Procedures.** Clinical activities were kept at a minimum, since the effort was directed primarily toward improving the MRR, to prevent any recurrence of the ocular complications seen in the first month of the fiscal year. Four patients in far advanced stages of the disease were treated by B<sup>10</sup> administration of 35 mg/kg body weight and thermal neutron exposure of from 100 to 200 sec. Skin flaps had been turned back on these patients. The brain cortex received from  $1.72 \times 10^{12}$  to  $1.21 \times 10^{13}$  neutrons/cm<sup>2</sup>. This marked increase in neutron exposure resulted in no immediate adverse effects.

Patients previously treated were observed and studied. For the first time phenomena of multiple tumors, both contra- and ipsilateral, were seen. Nonetheless, one of the patients survived for 18 months with minimum impairment and maximum effectiveness following a single treatment. This is the maximum longevity attained thus far in glioblastoma multiforme. The clinical evaluation of these results is being continued by means of many techniques, including serial sections of whole brain and tumor *in situ*, staining and histochemical techniques, and other suitable methodology. The first 16 cases are now being studied in collaboration with the Armed Forces Institute of Pathology. In one case, it is thought that radiation effects occurred in the neoplasm, in two other cases radiation effects were suspected, while in the remaining cases the neutron flux was presumably not sufficient to affect the deep-seated neoplasms. In an attempt to evaluate the effect of this therapy, a sarcoma of vascular origin in the cerebellum was studied. Extensive destruction of the tumor was demonstrated, but additional study is needed to determine whether any treatment-

induced changes occurred in the nonneoplastic structures.

Studies are continuing to elucidate the mechanism of action of neutron capture therapy. Selectivity seems to be founded on kinetics rather than on specific distribution or uptake. Studies of the pharmacology of boron compounds are also continuing. In addition, lithium-containing compounds and salts containing both lithium and boron are being investigated. Amelioration of the toxicity of these compounds by glucose is still being studied.

## ACCELERATOR RADIOLOGY

The study of particle effects that are or may be of medical significance is not limited to reactor radiology. Accelerators can provide "pure" energy spectra of a variety of particles and hence are used to extend and diversify the medical program.

New equipment had to be developed to permit the exposure of animals to a variable neutron dose with neutrons of known energy. Essentially monoenergetic beams were produced by a device in which a 2.8-Mev proton beam strikes a tritium target. This device, designed to assure uniform dose, was used in experiments on mice.

The relative biological effectiveness of lower energy neutrons was compared to that of neutrons of 2 Mev energy or higher. The effect of these particles on spermatogonia was such that depletion of spermatogonia is now believed to be the most sensitive biological index of radiation effects.

By using the 60-in. cyclotron a lesion was produced in the cerebral cortex of animals at a depth not exceeding 2 mm. Damage was produced in an isolated strip of cortical tissue not exceeding  $\approx 50$   $\mu$ , involving a layer of very few neurons.

The use of ionization chambers of tissue-equivalent plastic material was found necessary for the appropriate response correlation. It was indicated that with the bilateral exposure used the energy deposition throughout these artificial mice was uniform to at least  $\pm 10\%$ .

Accelerator-produced radioactive isotopes continued to be of interest. I<sup>124</sup> was used for labeling a globulin fraction in the study of protein metabolism in cancer. A new, cyclotron-produced isotope, Cu<sup>67</sup>, was used in a study of metal binding in mammals and in a study involving the role of ceruloplasmin in Wilson's disease.

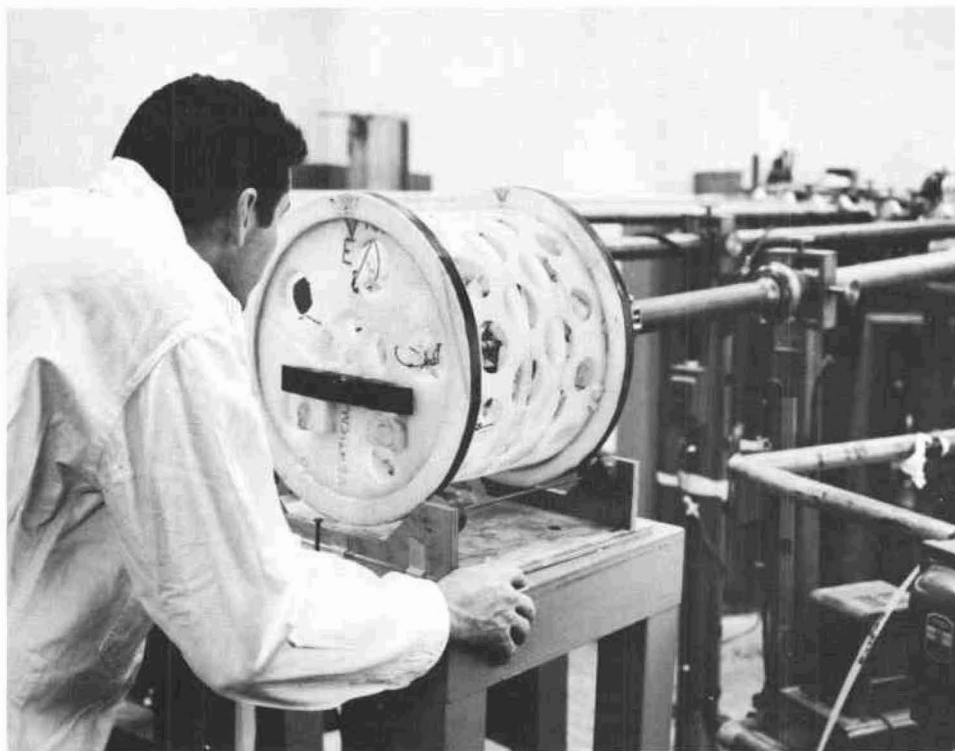


Figure 1. Alignment of mouse exposure cage with beam pipe of 3-Mev Van de Graaff generator (for monoenergetic neutron rbe studies).

### RADIOACTIVE TRACER METHODOLOGY

In principle, tracers make it possible to deduce the kinetic properties of steady-state systems by mathematical analysis of families of curves that might be representative of the behavior of the exchangeable substances in the system. In practice, this purely mathematical reasoning must be supported by analysis of experimental data to determine the best fits. For rapid analysis of such data it became necessary to evolve an analogue computer that made possible the simulation of the injection of constant initial charge or constant initial voltage, or of continuous injection of charge. With this device up to four curves may be displayed simultaneously. This instrument has been used in elucidating electrolyte exchanges in human beings and in other related investigations. Also evolved from data analyzed on the computer was a mathematical model interpreting the degradation of intravenously injected  $I^{131}$ -labeled proteins.

The kinetics of administered  $Sr^{85}$  were studied for as long as 5 mo in nine patients with fundamental disturbances in their calcium metabolism. The

turnover rates of  $Zn^{65}$  and  $Cs^{137}$  were also determined in four patients over long periods of time. These matters were studied in experimental animals also, and the dietary administration of stable strontium was found to decrease significantly (by 70%) the retention of injected radiostrontium by rat skeletal tissue.

The extrathyroidal metabolism of halides such as  $I^{131}$ ,  $I^{132}$ , and  $Cl^{38}$  were studied in the rat. By using *in vivo* gamma-ray counting, some of the animal experiments could be confirmed in a preliminary fashion on three patients. In both rat and man the gastric mucosa was found to excrete halides faster in the fed state than in fasting.

Studies on the kinetics of uptake and distribution of fission products in animals were continued during the year. The exchange capacity of the skeleton for  $Y^{89}$  was compared with that for previously studied  $Sr^{85}$ .

The kinetics of vitamin  $B_{12}$  administered intravenously to patients have indicated that plasma clearance and mixing of vitamin  $B_{12}$  go on simultaneously. The clearance curve consisted of two components with half-lives of a few minutes and



a few hours, respectively. The fecal and urinary excretion also showed two components, with half-lives of  $\approx 1$  and 300 days, respectively. The kinetics of labeled vitamin B<sub>12</sub> and the turnover of whole-body B<sub>12</sub> can be satisfactorily established only by using true tracer doses and a whole-body counter.

A transport mechanism which might be operating normally in plasma to transport manganese was studied during the year. It involves one of the beta globulin fractions of the serum proteins which utilizes manganese and oxygen as substrates. The incorporated element is trivalent in form, which indicates that it may be possible to reconcile the discrepancies between the *in vivo* and *in vitro* behavior of manganese and magnesium. *In vitro* magnesium and manganese are divalent and hence behave very much the same. *In vivo* magnesium remains divalent, but manganese becomes trivalent, and therein seems to lie the explanation of the sharp differentiation between these two elements, which is characteristic of integrated life.

The behavior of Cd<sup>109</sup>, as compared to that of the isotopes already discussed, was found to be unusual. While the isotopes of natural body constituents show characteristic movement and characteristic total-body turnover rates, Cd<sup>109</sup> showed almost no total-body turnover rate and very little movement from one organ to the next. Indeed, its motility was not affected by large loads of stable cadmium.

### NEUTRON ACTIVATION ANALYSIS

Analyses of small samples, such as needle biopsies, have always been difficult. In the case of trace metals occurring in very small concentrations within the body the analytical problems are inherently difficult. This is particularly so with analyses of serum, plasma, or blood, because in these compartments trace metals are even scarcer than they are intracellularly. Hence, exceedingly sensitive methods had to be devised.

At present the most sensitive method seems to be neutron activation analysis. As stated earlier, the approach consists of exposure of a sample to reactor neutrons which render radioactive various elements in the sample. This maneuver is, therefore, colligative. To separate and measure any one of the many elements, the procedure is either to sort out the radiations characteristic of each element, or to utilize a chemical separation of the

element in question. Both approaches have been used in the Medical Department during the year with considerable success. The first approach involves use of electronic circuitry designed to discriminate against "uninteresting" radiations emanating from an activated sample and to favor "interesting" radiations. This has been achieved in the determination of manganese, which is an ultramicro constituent of tissue. It has been possible to visualize radiomanganese while suppressing the energy peaks of sodium, potassium, chlorine, etc., that had become coactivated.

The second approach, that of chemical separation, has been developed into a clinically useful microprocedure. Previously analysis of plasma or serum required very large samples of these fluids, while with neutron activation analysis 0.1-ml samples of plasma or blood containing  $\approx 0.3 \mu\text{g}$  are sufficient for an accuracy of  $\pm 3\%$ .

The principle of activation analysis has contributed to the concept of a new brain center. The assumption is that gold thioglucose produces a lesion in the "satiety" center of the brain, since a permanent tendency to overeat was induced in animals injected with gold thioglucose. Thermal neutron activation of gold and sulfur in brain slices from such animals showed the location and other structural features of the satiety center.

### METABOLISM AND HOMEOSTASIS

A highly radioactive form of the hormone vasopressin was synthesized. This permitted the elucidation of facts concerning the functional attachment of the hormone to its receptors. The chemical information proved to be crucial in interpreting the hormone's physiological action. In turn, this led to a study of hypertensive phenomena in animals and man, since vasopressin has long been vaguely implicated in such phenomena.

The evidence pertaining to vasopressin has suggested a similar approach to the study of insulin and other hormones containing S—S linkages in their molecules. This novel approach has shown promise in preliminary experimentation with radioactive hormones.

Mn<sup>54</sup> was shown to become incorporated into the heme fraction of human hemoglobin. Feeding of stable manganese elevated the metal's concentration in erythrocytes without depressing the hemoglobin content. These data indicate the existence of a manganese-containing human hemo-



Figure 2. Collection and measurement of respired  $C^{14}O_2$  in studies of carbohydrate oxidation in diabetic patients.

globin, and its role is being investigated vigorously. This type of experiment was strengthened by studies of the hormonal control of manganese which showed a specific action of glucocorticoids on this element's distribution in mice. Furthermore, it has been found that manganese is transported by a beta globulin of human serum, which binds its trivalent form *in vitro*. The valence of this element is interesting, since the same valence has been implicated in reactions of manganese with some phenothiazine drugs.

Studies with radioactive sodium have revealed consistent differences between normotensive and hypertensive humans with regard to their sodium metabolism. These differences were reflected also in animals rendered experimentally hypertensive.

Studies with  $C^{14}$ -labeled compounds of ketogenesis in man have been too extensive and too productive for summarization here. They have covered both mature and juvenile diabetics and have resulted in considerable insight into both the biochemistry and pharmacology of diabetes. These studies utilized and extended the knowledge of glucose formation in man already extensively documented at the BNL Medical Research Center. In

view of the cardinal role of bicarbonate in all aspects of diabetes, various radioactive forms of this substance were extensively studied on the basis of concepts evolved at this Laboratory. Detailed insight into the metabolic pathways of glucose, its precursors, and its breakdown products has been gained. The regulation of lipid and cholesterol metabolism by hormones was also studied with similar techniques.

Work on the metabolism of *Trichinella spiralis* was extended and amplified. This work has utilized techniques kindred to those employed in the study of tryptophan, its precursors, and its breakdown products. The latter experiments revealed significant differences between normalcy and neoplastic states.

Studies of the turnover rate of proteins in patients have been greatly assisted by the use of total-body counting techniques. Emphasis was placed on long-term observations because of the fruitfulness of this approach. Studies of multiple myeloma patients have revealed significant metabolic differences between various types of the disease as regards the metabolism of normal and aberrant proteins.

MEDICAL RESEARCH  
CENTER  
WHOLE-BODY COUNTING  
FACILITY

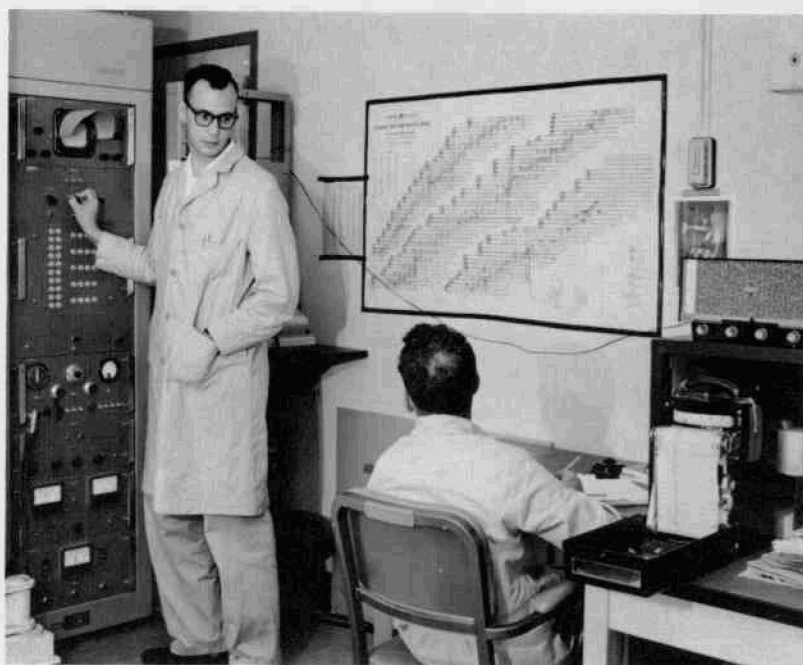


Figure 3. Personnel operating 100-channel analyzer and print-out equipment during measurement of patient in whole-body counter.

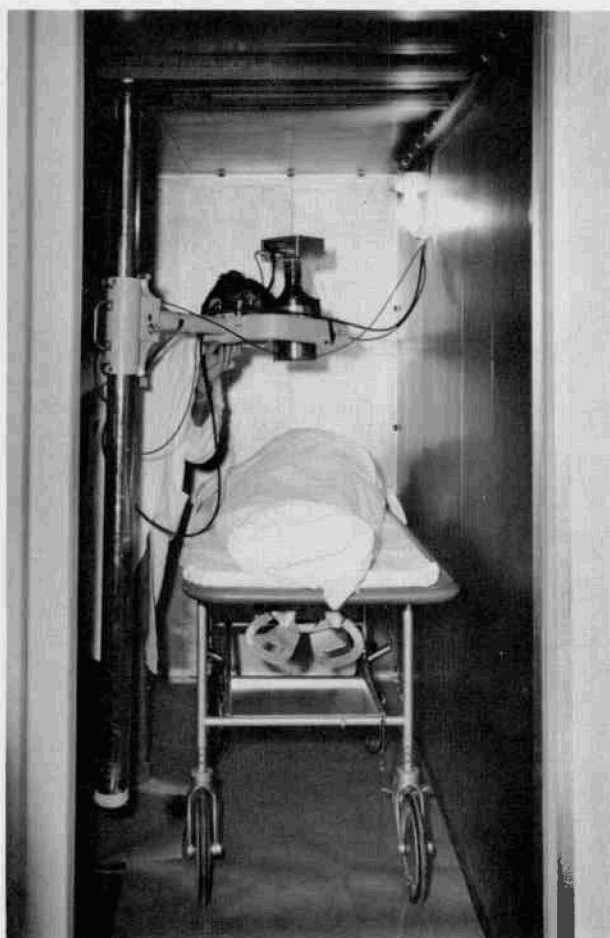


Figure 4. Detector crystal is positioned over a patient who has received neutron capture therapy in treatment of brain cancer. A count of the neutron-induced gamma activity will provide a measure of the intensity of neutron exposure.



Figure 5 (Top, left). Plastic phantom, placed in standard measuring position, is positioned under a detecting crystal of thallium-activated sodium iodide.

Figure 6 (Top, right). Use of the whole-body counter in the study of protein metabolism in cancer. An iodine-labeled protein fraction has been given to this patient with multiple myeloma; its disappearance can be followed accurately over a period of weeks.



Figure 7. Plastic phantom of standard man, filled with radioactive solution, is carried into whole-body counter. Measurements on phantoms provide data for calibration and comparison of data obtained from patients.



Figure 8 (Top, left). Brookhaven employee, seated in whole-body counter for standard measurement during a survey of employees, receives instructions on the use of the two-way intercommunication system,

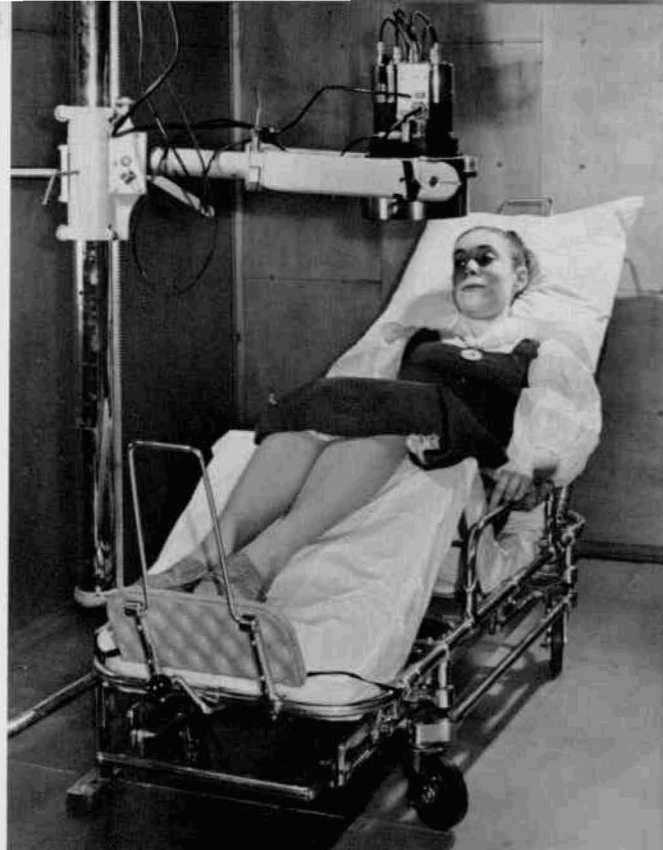


Figure 9 (Top, right). Patient seated in whole-body counter for measurement of radioactive sodium in study of management of hypertension.

Figure 10. Crystal detector is positioned over a patient from the Industrial Medicine Clinic who may have been exposed to internal radioactive contamination. By whole-body measurement, the presence and extent of such contamination can be determined.





Long-term observations of the type alluded to above have been found fruitful in studies of Co<sup>60</sup>-labeled vitamin B<sub>12</sub>. This important problem is being studied in a number of persons with various disease conditions.

Work has continued on the metabolism of ribose and deoxyribose nucleic acids (RNA and DNA) with the aid of tritiated compounds. Tritiated cytidine was incorporated into cell nuclei, and the time sequences of this incorporation were compared with those resulting from studies with other compounds. These experiments supported the idea that RNA synthesis begins in the nucleus.

The metabolic fate of tritiated thymidine was studied in patients with hemopoietic equilibrium. This permitted detailed assessment of the compartments participating in the proliferating processes of the hemopoietic system. Hence, the observations made previously on blood diseases have become amenable to accurate interpretation.

It was discovered that mice could be made immunologically tolerant to bovine serum albumin by being given this antigen soon after birth. The long duration of this tolerance indicated prolonged persistence of the antigen at intracellular sites. In studies with an antigen plus an antibody, higher immunity levels were produced than with either agent alone. It was also found that antigen can be degraded at an accelerated rate under conditions of antigen excess with regard to antibody. These findings may be of assistance in the development of effective immunization by the use of antigen-antibody mixtures.

#### CELLULAR REPRODUCTION, MIGRATION, AND TRANSFORMATION

Deoxyuridine was labeled with I<sup>131</sup> in order to study its incorporation into DNA in the intact man. Three patients with neoplasms have been studied with this compound, and the data are being reviewed to determine any effects upon the rate of cell proliferation in the neoplastic tissues.

The studies with tritiated thymidine have continued. An *in vitro* technique was developed which permits estimation of the proliferative potential of individual cell types. A series of findings seems to throw doubt on the accepted hypotheses that neoplastic tissues have high turnover rates and that chemotherapy affects such processes. By the same token, these and related findings seem to delineate new hypotheses concerning malignant proliferation.

A detailed study was completed in which cytidine, uridine, and thymidine, all tritiated, were used as precursors to the synthesis of DNA. It confirmed a previously reported sequence of labeling from the nucleus to the cytoplasm as indicating a shift of RNA. The results suggested also that there might exist a DNA precursor more intricate than a mononucleotide, in a closed system of precursors participating in DNA synthesis. The closed precursor system had a constant turnover time and a unidirectional flow to DNA.

While these latter experiments were done on HeLa cells, hemopoietic tissue was also studied with similar methodology. Furthermore, human lymphosarcoma was submitted to quantitative studies of its RNA turnover. These showed that lymphosarcoma is not a rapidly proliferating tumor, contrary to established belief.

The above studies and others of a similar nature culminated in an investigation of malignant diseases of humans. Various forms of malignancy were studied by means of determinations of thymidine incorporation into DNA. Up to the present writing it appears that, in myeloma at least, only a small portion of the myeloma cells retain their capacity to propagate the tumor, while the balance reach a mature state and remain as parasites to the host organism. These observations appear amenable to generalization, since they might apply also to the leukemias and to solid tumors currently under investigation.

The techniques alluded to above have necessitated detailed studies of the absorption, incorporation, and testing of the tritiated compounds. Furthermore, correlations had to be drawn with older methods (such as the mitotic index) in healthy as well as irradiated humans.

These studies have also been extended to problems involving antibody synthesis as well as the synthesis of DNA in embryo and adult mice, in osteoclasts of bone, in fracture repair, and in peripheral blood cells. The study of the effect of aging on fracture repair and osteoclastic activity was supplemented by extensive histochemical and autoradiographic observations.

#### ENVIRONMENT AND RADIATION

In a long-term study of the radioactive burden in the bodies of Laboratory workers, begun this year, 130 staff members were subjected to measurements by means of total-body counting tech-

niques. Levels of  $K^{40}$ ,  $Cs^{137}$ , and  $Zn^{65}$  were quantitated.

The effort to study maneuvers capable of yielding radiation protection has been vigorously continued. The possibility that barium meals protect the gastrointestinal tract during diagnostic exposure received experimental support. The bleeding tendency of irradiated dogs could be followed by observing the red cell output in thoracic duct lymph. Transfusion of intact platelets dramatically decreased this bleeding, while disintegrated platelets reduced only the prothrombin consumption. Lyophilized platelets were unsatisfactory.

A detailed analysis of human lethal dose of radiation, undertaken from both the military and civilian standpoints, indicates that the  $LD_{50}$  for man is probably below and certainly not above 400 to 450 r given as total-body radiation.

Among materials considered capable of radiation damage, Thorotrast and technetium were assessed. The first was investigated in persons who had received Thorotrast diagnostically. The latter was studied in animals, where it was found to concentrate in thyroid and in eggs, among other sites. The possibility of retardation of growth following exposure to atomic bomb and fallout radiation is being studied in rats. Similarly, the radiosensitivity of immune responses, both primary and secondary, has been investigated. The results of this work showed a surprising dissociation between primary and secondary immune responses; the former were considerably more radiosensitive than the latter.

Tritium and tritiated compounds were studied extensively by several groups with respect to their radiation effects, in view of the cardinal role these compounds are now playing in investigation, diagnosis, and evaluation of therapy. Evidence too extensive to summarize has been gathered which seems to have quantitated the radiation effects on various end organs. Dosimetric studies have been correlated with biological responses. The experience gained not merely is applicable to the utilization of tritiated compounds but, more importantly, has provided extensive insight into mechanisms involved in tissue damage by radiation. Intracellular, as opposed to cellular, irradiation can now be accomplished in mammals. This technique is being evaluated with regard to its possible modification of phenomena observed by means of labeling nuclei with tritiated compounds as well as with regard to its obvious therapeutic implications.

Studies of carcinogenesis following radiation exposure are continuing. A maximal neoplastic response of mammary tissues may be induced in female rats with 400 r of 250-kvp x-rays. Interactions between carcinogenesis and pregnancy were discovered: pregnancy aggravated the rate of mammary tumor growth. Hyper- and hypothyroidism were without measurable carcinogenic effect. Marked variations between rat strains were noted in these studies. Removal or shielding of the ovaries reduced carcinogenesis.

The role of the ovary with respect to carcinogenesis in the breast was investigated further. A similar study was done with regard to the testes. The experiments with solid tumors were extended to leukemogenesis.

### OTHER BIOMEDICAL STUDIES

Gasometric procedures were developed permitting accurate analyses of gases present or generated in 0.1 ml of blood, plasma, or other biological fluids. These procedures include determination of anesthetic gases, of microamounts of  $C^{14}O_2$ , etc. The method of determining protein-bound iodine in serum was improved by the use of  $I^{131}$ . Various interactions between plasma proteins and lipids were studied by isotopic and other physical techniques. Bacterial enzymes have been utilized in studying the linkage between carbohydrate and protein in mucoproteins.

The transfer of sugars through cell membranes was studied further. A partial isolation of the transfer system was achieved. Several structure-activity relationships were elicited in studies of inhibitors of the transfer system. Phlorizin analogues were particularly useful in the study of inhibition of sugar transport.

### HOSPITAL

The increase in the use of hospital facilities for research studies continued during fiscal 1960, as shown by 9021 patient days compared to 8804 for the previous year. Of the 117 persons admitted, 5 were employees or employees' dependents, and 112 were research patients. Outpatient visits increased to 968 from 751 for the previous fiscal year; and the hospital ambulatory service for patients who could be managed satisfactorily in this manner to reduce hospitalization time and for those whose diagnostic and therapeutic tests could be completed without more costly hospitalization



accounted for an additional 67 persons seen on the research service.

Sixteen radioactive isotopes of 13 different elements were used in diagnosis and therapy of 69 patients. The major disease states so studied and treated in the hospital were glioblastoma multiforme, leukemia, multiple myeloma, motor disabilities of the central nervous system, hypertension, disorders of the thyroid, arthritic diseases, polycythemia vera, and obesity.

### **INDUSTRIAL MEDICINE**

With only one Industrial Medicine physician during nine months of fiscal 1960, visits dropped to 13,650 from 15,157 for the previous year. Routine annual examinations continue to be scheduled at  $>12$ -mo intervals because of time required daily for loss-of-time prevention visits and the priority scheduling of examinations of employees engaged in radiation areas or in hazardous activities.

Employees whose urinalyses show radioactivity, as well as those who may have acquired body burdens of radioactive substances, are now being routinely examined in the whole-body counting room, for low level radioisotopic contamination. Scheduling of this group of employees has been given priority over use of the counter for research activities, which, in turn, has developed a well-defined requirement for a second whole-body counting unit.

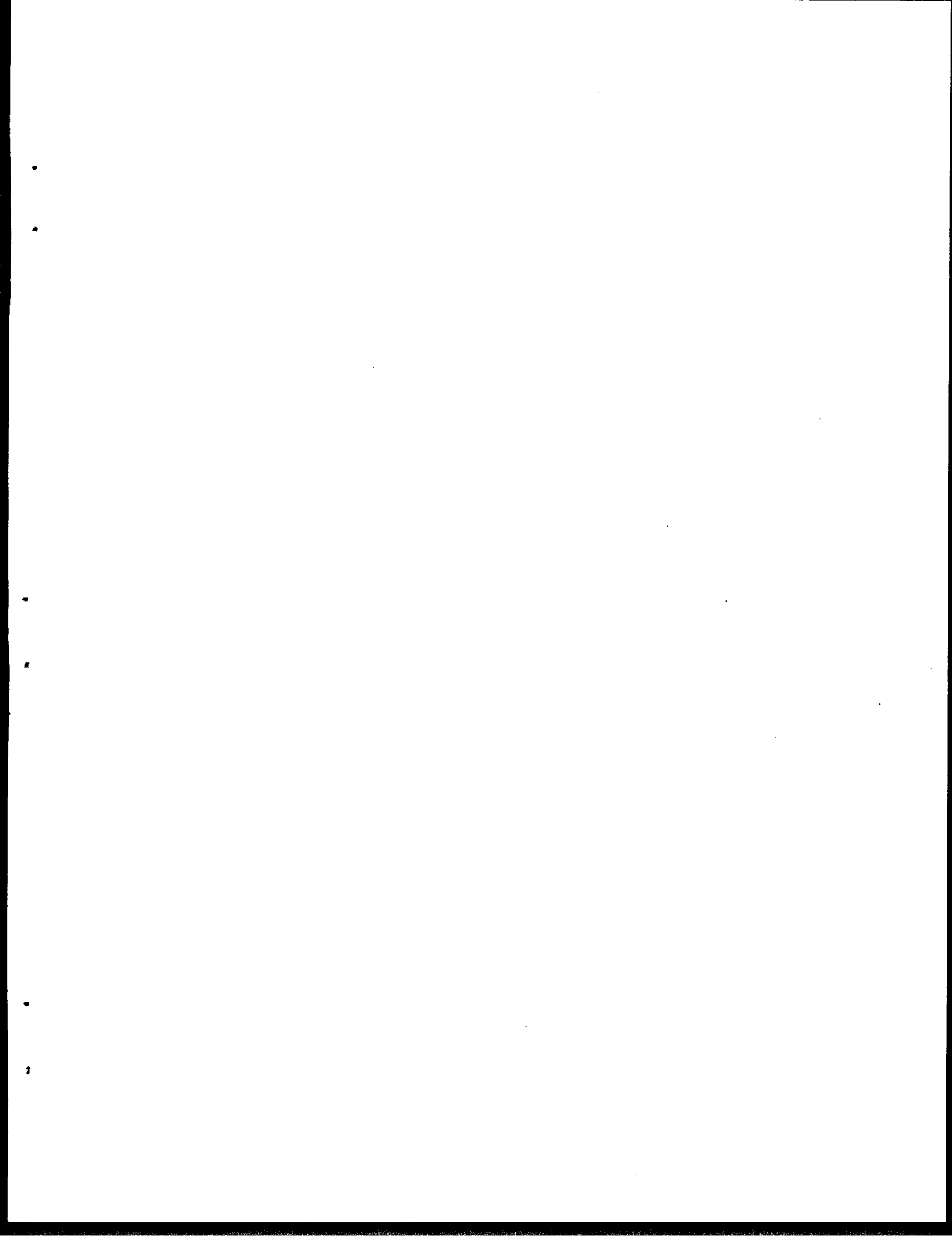
The decrease in the number of x-ray examinations to 1938 from 2236 during the previous year reflects the drop in routine physical examinations. In general, the state of physical fitness of BNL employees continued to be excellent in relation to the tasks to which they are assigned.

### **SPECIAL PROJECTS**

Pending completion of full agreements with the Trust Territory of the Pacific Islands with regard to the scope and intensity of surveys to be carried out in the future, a brief interim scanning survey of the Marshallese people accidentally irradiated by fallout in 1954 was carried out by two members of the Medical Department.

The fourth in a series of annual conclaves on nuclear science in medicine sponsored jointly by the Division of Biology and Medicine of the US AEC and the Medical Department was held at the Medical Research Center. Concerned with "Nuclear Medicine in Surgical Research and Practice" the two-day program was participated in by 66 Surgical Department chairmen from medical schools in the United States and Canada.

The first Medical Student Institute of Nuclear Medicine was conducted over a five-week period early in the fiscal year. Eight medical students and three faculty members of the University of Texas participated.





# 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 Graphite Research Reactor was operated 82.2% of the year. The routine operating schedule was interrupted on 16 occasions for a variety of causes. There were 5 emergency shutdowns caused by electric power failures; electronic failures within the instrumentation and control system caused 6 accidental shutdowns; snow plugged the inlet air filters on 2 occasions; and human operating errors accounted for 3 shutdowns. There were no reactivity incidents or cases of serious misoperation.

The maximum power of the pile was increased from 17 Mw to 19 to 20 Mw, and the size of the loading was increased from 479 to 548 channels. The maximum thermal neutron flux remained at  $\approx 2 \times 10^{13}$  neutrons/cm<sup>2</sup>-sec.

In order to maintain reactivity requirements, 69 new fuel channels were added and 710 elements that had approached  $\approx 40\%$  burn-up were replaced. The average burn-up of all elements removed from the reactor, including several that had been accidentally blown out of channels by the cooling air, was 36.7%.

### Graphite Annealing

The graphite moderator structure was annealed 3 times during the report period, bringing the total number of annealing operations to 13. Since the tenth anneal, the annealing periods have been 16 hr each at graphite temperatures between 300° and 370°C, and in every case the recovery of growth since the previous anneal was  $> 100\%$ . The effect of the longer annealing period seems to have been to increase the shrinkage. A plot of the structure growth vs nvt, shown in Figure 1, illustrates the effect of annealing on growth for each of the 13 annealings.

### New Cooling Fans

As a result of reloading the reactor with enriched fuel elements, the volume and pressure loss

requirements for the cooling air were reduced sufficiently to warrant the replacement of 3 of the 5 primary fans with fans designed to meet the new specific requirements of the system. While the new fans were being installed (from March through June), the reactor power was reduced somewhat at times, but it was not necessary to shut down the reactor, since the No. 4 and No. 5 fans were available. Savings of electric power due to operating with the 3 new fans are expected to equal their cost in  $< 2$  yr.

### Utilization of the Graphite Research Reactor

With the exception of those on top, the reactor's experimental facilities have been, for all practical purposes, fully used. The shielding facility on top of the reactor was used for a few minor experiments. The medical facility on top of the reactor has been used only a few times since the Medical Research Reactor became available.

Space in the reactor was made available to outside organizations to the extent that no interference with Brookhaven experimental programs was encountered. A list is given below of the outside organizations making significant use of the reactor beyond simple service irradiations.

**Naval Research Laboratory.** Steel specimens were irradiated at elevated temperatures and were then sent to NRL for studies of radiation damage. This work was done under Government Contract No. NRL/AEC 5-60.

Magnetic core materials were irradiated to determine the effect of reactor irradiation on the magnetic properties of various materials. This work was done under Government Contract No. NRL/AEC 5-60.

**Rensselaer Polytechnic Institute.** Experiments were carried out to determine the effect of fission fragments on circulating oxygen-nitrogen mixtures through a tube containing quartz fibers that were 15 wt % U<sub>3</sub>O<sub>8</sub> (90% enriched in U<sup>235</sup>). This work was performed under Government Contract No. AT-30-3-321.

**Bell Telephone Laboratories.** Studies were carried out on neutron diffraction from crystals at room temperature and at 4.2° K. This work was done under a utilization agreement.

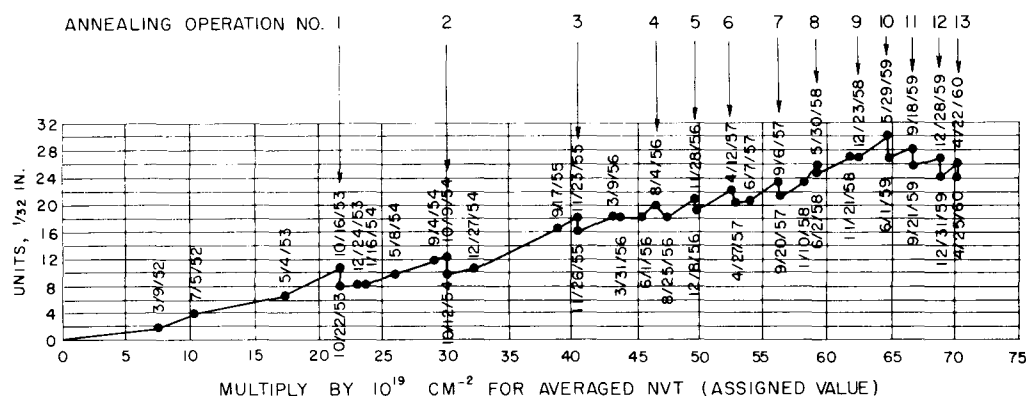


Figure 1. Maximum vertical graphite growth (near gap) in Hole B-O-15 versus averaged exposure.

Irradiations were made to study radiation damage in semiconductor materials and devices. This work was done under Government Contract No. AF-33-616-6235.

**General Electric Company.** Studies of magnetic and crystal structure were made from neutron diffraction patterns obtained from various crystals and powders at different temperatures. This work was done under a utilization agreement.

**Westinghouse Electric Corporation.** In-pile measurements were made of the effects of reactor irradiation on the thermoelectric parameters of some materials of interest for application as thermoelectric materials. This work was done under Government Contract No. AT-30-3-500.

**Sperry Gyroscope Company.** Measurements were made of changes in the electrical properties of certain electronic components and plastic materials while an in-pile irradiation was in progress. This work was done under Government Contract No. AF-33-600-39801.

Changes in the magnetic and electrical properties of some magnetic core materials were measured while irradiation was in progress. This work was done under Government Contract No. AF-33-600-39801.

**P.R. Mallory and Company.** Certain types of precision resistors were irradiated at elevated temperatures to determine the changes in the electrical properties due to radiation damage. The measurements were made during the progress of the irradiation. This work was carried out under Government Contract No. AF-33-616-6164.

**Markite Company.** A furnace containing potentiometer parts together with monitoring foils was installed in an experimental hole and meas-

urements were made to determine changes in the electrical properties of these elements due to radiation damage, both at ambient temperature and at 500°C. The duration of the test was  $\approx 1200$  hr. This work was done under Government Contract No. AF-33-616-6709.

**Anton Electronics Laboratories.** Performance tests and measurements were made on an ionization chamber, a BF<sub>3</sub> counter, a boron-coated counter, and a fission chamber in a thermal neutron flux of  $10^{11}$  neutrons/cm<sup>2</sup>-sec. These detectors together with their connecting cables were being tested for possible use in the Enrico Fermi Reactor.

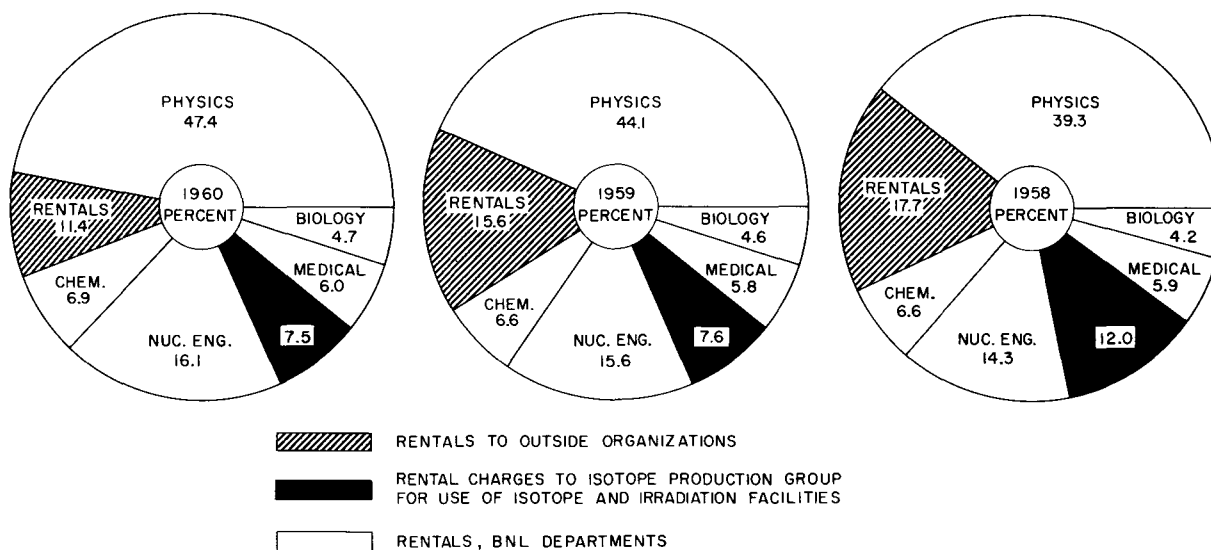
The following organizations carried out experiments at the Graphite Research Reactor under an internal arrangement with the BNL Physics Department.

**Columbia University.** In the helium recoil experiment measurements were made of the energy spectrum of recoil nuclei which have emitted a beta-particle of discrete energy in the decay of He<sup>6</sup>. The He<sup>6</sup> is produced according to the reaction Be<sup>9</sup>(n,γ)He<sup>6</sup>, by bombarding beryllium hydroxide in the reactor.

A single-crystal neutron spectrometer with a neutron energy range from 0.0006 to 10 eV was used to measure total neutron cross sections. The cross sections were determined for samples of boron of various enrichments, manganese compounds, ammonium halides, and benzene compounds formed by the addition of methyl groups.

**Naval Ordnance Laboratory.** The powder diffractometer was used jointly with the BNL Solid State Physics Group for several powder diffraction experiments, mostly on magnetic materials. Some were performed at liquid helium temperature.

Table 1  
Summary of Reactor Use Charges



	Fiscal year					
	1960		1959		1958	
	<u>Rentals</u>					
Outside Organizations	\$ 149,119		\$ 175,294		\$ 197,562	
BNL Departments						
Physics	\$621,827		\$494,694		\$439,898	
Chemistry	90,480		74,280		74,280	
Nuclear Engineering	211,960		175,020		159,420	
Reactor Isotope Group	97,920		85,408		133,874	
Medical	79,200		66,000		66,000	
Biology	62,240	1,163,627	51,790	947,192	46,754	920,226
Total	\$1,312,746		\$1,122,486		\$1,117,788	
	<u>Reactor Irradiations</u>					
Outside Organizations*	\$ 97,920		\$ 81,600		\$ 83,400	
BNL Departments						
Physics	\$ 17,730		\$ 16,427		\$ 12,121	
Chemistry	8,534		4,553		9,114	
Nuclear Engineering	11,234		16,744		37,620	
Medical	3,934		6,489		4,973	
Biology	218		362		3,474	
Instrumentation and Health Physics	2,380					
Reactor		44,030	280	44,855	1,648	68,950
Total	\$ 141,950		\$ 126,455		\$ 152,350	
Total Use Charges	\$1,454,696		\$1,248,941		\$1,270,138	

\*Since Table 2 indicates income from sale of these items, the numbers are not the same.

**Picatinny Arsenal.** The water-cooled facility was used primarily for the irradiation of special materials at less than ambient temperatures.

The use made of the reactor by the BNL research departments is discussed in the appropriate sections of this report. Table 1 illustrates the relative use of the reactor during the past three years by the various departments and outside research organizations. Use of the reactor for isotopes and service irradiations is also indicated in this table.

An increase in reactor rental rates of  $\approx 20\%$  became effective on July 1, 1959, for BNL departments. For outside organizations, the change was made effective as soon thereafter as practical under the various working arrangements. The increase in rentals for 1960 over 1959 is primarily due to this change in rental rates.

### Procurement of Special Materials

The procurement for the scientific departments of all radioactive and stable isotopes as well as special materials controlled by the AEC is a responsibility of the Isotopes and Special Materials Group. In this connection, 209 purchase orders were placed for radioisotopes, 41 for stable isotopes, and 53 for special materials. Against these orders,  $\approx 408$  shipments were received and processed. These figures represent an increase of  $\approx 20\%$  over the previous year. Of the orders placed,  $\approx 26\%$  were for the Medical Department, 21% for the Biology Department, 15% for the Physics Department, 18% for the Nuclear Engineering Department, 16% for the Chemistry Department, and 4% for the Health Physics Division.

The Isotopes and Special Materials Group is also responsible for annual inventories of radium sources and purchased stable isotopes and for negotiations for the loan of valuable stable isotopes. The number of radium sources at BNL was reduced from 38 to 34 upon write-off of 4 small sources to waste disposal, but the total activity of 5.86 curies and value of \$38,876.00 remain essentially the same as last year. The inventory of purchased stable isotopes is valued at \$26,851.00, slightly higher than last year. During the year, 7 new loan agreements were made for stable isotopes, which brings the total number of loans to 34 with a value of \$146,895.00.

Three new irradiation programs were initiated at the National Reactor Testing Station, Idaho Falls, Idaho. The first, for the Chemistry Department, is a one-year irradiation of enriched  $\text{Pb}^{204}$

at a thermal neutron flux of  $4.3 \times 10^{14}$  neutrons/cm<sup>2</sup>-sec to produce  $\text{Pb}^{205}$ , which has a  $5 \times 10^7$ -yr half-life. The second program, for the Physics Department, is the irradiation of samples of diamond powder by fast neutron flux dosages of  $2 \times 10^{20}$ ,  $5 \times 10^{20}$ , and  $8 \times 10^{20}$  neutrons/cm<sup>2</sup>-sec for studies of radiation damage. The third program involves the testing of prototype fuel elements for the Brookhaven High Flux Beam Research Reactor. The program for production of 500,000 curies of  $\text{Co}^{60}$ , in progress for two years, is continuing, with delivery expected soon of  $\approx 90,000$  curies of  $\text{Co}^{60}$  which were discharged from the Engineering Test Reactor at the end of the fiscal year.

### Isotope Production and Irradiation Services

Table 2 presents a three-year summary of the volume and income involved in providing irradiation services and processed radioisotopes to outside organizations. Although the number of irradiations has remained relatively stable, the large variations in income have resulted from the sporadic sales of kilocurie  $\text{Co}^{60}$  sources of the BNL type. The five  $\text{Co}^{60}$  sources shipped in fiscal 1960 totaled 8752 curies, compared to  $\approx 10,000$  in 1959 and  $\approx 12,000$  in 1958. Of the five sources shipped this year, two, totaling 5413 curies, were supplied on a no-charge basis.

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 this subsidy, borne by Brookhaven, has risen constantly, as shown in Table 2.

The use of the BNL 60-in. cyclotron for irradiations for outside organizations has always been limited. The cyclotron irradiations listed in Table 2 were done as part of cooperative programs with universities or for federal agencies.

Table 3 shows the minor variations in the distribution of customers and shipments over the past three fiscal years. The shipments of special radioisotopes supplied by the Hot Laboratory are shown in Table 4.

At the request of the AEC, a detailed analysis was made of reactor operation costs to determine the full costs to be borne by the various reactor facilities and irradiation services. As a result of this analysis, new rates have been proposed to insure full cost recovery. The facility rental rates are relatively unchanged, since an increase in these rates of  $\approx 20\%$  was put into effect on July 1, 1959. The



Table 2

## Three-Year Summary of Irradiation Services to Outside Users

	Fiscal 1960		Fiscal 1959		Fiscal 1958	
	Number	Volume, \$	Number	Volume, \$	Number	Volume, \$
Reactor irradiations	1,025	54,294	1,189	73,507	1,153	66,683
Processed radioisotopes	325	51,133	291	37,655	211	32,021
Research subsidy	—	(33,183)	—	(24,049)	—	(19,649)
Co <sup>60</sup> sources	5	8,186	12	42,320	8	29,753
Cyclotron irradiations	22	2,854	5	1,070	14	2,836
Total	1,377	116,467	1,497	154,552	1,386	131,293

Table 3

## Types of Customers Receiving Shipments

Type	Fiscal 1960		Fiscal 1959		Fiscal 1958	
	% of Customers	% of Shipments	% of Customers	% of Shipments	% of Customers	% of Shipments
Hospitals	18	40	25	43	10	35
Industrial	42	31	42	31	60	35
Universities	24	20	18	18	20	20
Government	7	5	10	5	10	10
Foreign	9	4	5	3	—	—

Table 4

## Shipments of Processed Radioisotopes

Radioisotope	Fiscal 1960		Fiscal 1959		Fiscal 1958	
	No. of shipments	Activity, mC	No. of shipments	Activity, mC	No. of shipments	Activity, mC
I <sup>132</sup>	70	1,359	48	3,900	45	2,020
I <sup>133</sup>	67	2,535	55	2,420	30	1,030
Mg <sup>28</sup>	158	556	181	23	130	20
F <sup>18</sup>	3	—	7	70	6	80
Y <sup>90</sup>	25	15,030	—	—	—	—
Ar <sup>38</sup>	2	—	—	—	—	—

service irradiation rates and handling charges have been increased by a factor of 2 to 4. The accounting for irradiation services has been revised and will yield a more detailed picture of cost versus income in three different areas in the next fiscal year.

A new brochure has been issued, entitled *Graphite Research Reactor, Facilities and Services Guide*, to replace the August 1956 version. Since that date, the reactor fuel loading has been changed from natural to enriched uranium, and changes have been made in many of the facilities. Revision of the brochure to incorporate these changes was de-

ferred until the new fuel loading approached equilibrium and data were available on the new neutron flux distributions.

#### Source and Special Nuclear Materials Accountability

Table 5 presents a three-year summary of the amounts of source and special (SS) nuclear materials on hand at BNL at the end of each fiscal year. The large decrease in natural uranium from 54,127 kg in fiscal 1957 is the result of its transfer to the depleted uranium category after irradiation as reactor fuel. The decrease in the amount of

depleted uranium reflects continuing shipment of this irradiated fuel to Oak Ridge National Laboratory for reprocessing. The  $\text{Pu}^{239}$  inventory has also been reduced by these shipments since it is present in the irradiated fuel. The  $\text{U}^{235}$  enriched to  $>75\%$  consists mostly of the present reactor fuel inventory, which has remained constant since fiscal 1958. The amount of  $\text{U}^{235}$  enriched to  $<75\%$ , however, has increased each year as a result of the increased activity and requirements of the Reactor Physics Experimental Group in the Nuclear Engineering Department. The large increase in the heavy water inventory in the past two years reflects the requirements of the Medical Research Reactor and the new High Flux Beam Research Reactor. The amounts of  $\text{U}^{233}$  and thorium, which are used in continuing research programs, remain essentially unchanged. Based on dollar values published by the AEC, the current inventory of the materials listed in Table 5 represents a value of  $\approx \$7,000,000$ .

As of January 29, 1960, calculations of burn-up of the reactor fuel loading had been brought up to date on the Remington Rand 409 machine. No further calculations were made on this machine, since the program was being revised for the IBM 704 tape computer. The reprogramming, however, proved to be more difficult than anticipated for an internally programmed machine; consequently, calculations of reactor fuel burn-up have been delayed for several months. Once the program is operating, the status of the reactor fuel will be computed after each reactor shutdown rather than every three or four months, as was the case with the Remington Rand 409 program.

A revision of *Procedures for Handling S/F Materials* was issued on April 1, 1960, under the title, *Accountability for Source and Special Nuclear Materials*. The revision reflects changes in Laboratory and AEC policies and procedures concerning the accountability of such materials and has been expanded in the area of controls on criticality problems. Some of the descriptive information formerly included has been deleted and is now incorporated in the *Special Nuclear Materials Facilities Manual* soon to be published.

To alleviate further the congestion in the fuel storage vault, a new area was established in the basement of the west reactor wing for interim storage of waste and scrap SS nuclear materials prior to disposal and for storage of many small stock items. In this way, the reactor storage vault

Table 5  
Source and Special Nuclear Materials on Hand  
at End of Year (in kilograms)

	Fiscal year		
	1960	1959	1958
Natural U	2,895	3,096	9,810
Depleted U	14,538	33,847	45,955
$\text{U}^{235} >75\%$	163	174	163
$\text{U}^{235} <75\%$	11,850	6,766	4,776
$\text{U}^{233}$	0.051	0.051	0.104
$\text{Pu}^{239}$	7	14	21
Th	254	259	194
Heavy water	13,321	8,879	176

can be organized for more efficient storage and criticality control of fuel elements and other fissionable materials.

As an AEC contractor, the Laboratory is required to submit monthly inventories of the listed materials, and both the materials and records are subject to an annual audit by the AEC. Because of prior commitments and a full schedule, however, the AEC did not perform such an audit at Brookhaven this year.

#### Medical Research Reactor

The Medical Research Reactor is operated by the Reactor Division as a service to the Medical Department. This reactor was operated on 151 occasions during the year and accumulated 196,261 kw-hr of operation.

The approved operating power level of 3 Mw was extended to provide for operation at 5 Mw for periods of time not to exceed 10 min. The excess reactivity allowable was extended to permit continuous operation at 1 Mw or periodic 8-hr runs at 3 Mw.

No significant operating difficulties have been encountered with this reactor.

#### HOT LABORATORY OPERATIONS

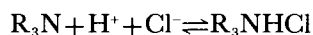
In addition to the production and processing of special isotopes and the performance of gamma irradiations, the Hot Laboratory staff provides many other services, some developmental in character, which are summarized briefly here.

#### Radiochemical Analysis

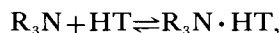
Chemical analyses for radioactive and/or stable constituents are performed on a service basis on all

types of radioactive materials. Both fundamental and applied research are also conducted in addition to and in support of this service work.

Of particular interest is a study undertaken to explain the unexpected observation that the distribution coefficient for thorium between an aqueous phase and an organic phase consisting of a mixture of triisooctylamine (TIOA) and thenoyltrifluoroacetone (TTA) in an inert diluent is greater by a factor of  $\approx 5$  than that obtained when TIOA is omitted. In investigating this fundamental problem, the hydrolysis reaction for tri-*n*-octylamine (TNOA) was studied, and the postulated mechanism



was verified. The equilibrium constant for this reaction was found to be  $1.3 \times 10^4$  in aqueous media, whether the solution contains only HCl or LiCl in quantities up to 2 *M*. The reaction between TNOA and TTA has also been investigated and has been determined to be



where HT stands for TTA. The formation constant for this acid salt was determined to be  $(1.43 \pm 0.10) \times 10^3$ . A spectrophotometric method was applied in determining this mechanism and the equilibrium constant. The study covered a 5000-fold range in amine concentration.

An investigation was begun recently of the applicability of contact radiography to the identification of active electrode sites at which electron transfer processes may selectively occur. Long-range objectives include the elucidation of the mechanism of electrode reactions and the refinement of voltametry of solid electrodes into an accurate nonempirical method of quantitative analysis. Thus far,  $Tl^{204}$  tracer gave well-defined contact radiograms of deposits having average thicknesses ranging from 10 to 1500 Å.

The following is a partial list of other problems investigated:

(1) A systematic effort has been made to apply the present incremental polarograph to various analytical problems, in particular the investigation of molybdenum, Mo-U mixtures, and Bi-U mixtures. For example, although the polarography of molybdenum is quite complex, incremental polarographic resolution between uranium and 100-fold excess molybdenum is well defined.

(2) A conventional polarographic determination of uranium in the presence of iron for application to plutonium-containing media has been devised.

(3) The investigation of the chronopotentiometric behavior of metal ions in fused LiCl-KCl eutectic continues. Polarograms of  $Pt^{+3}$  with a platinum needle electrode and a  $Pt^{+2}/Pt$  reference electrode have been obtained, and the diffusion current has been found to be proportional to concentration. When a liquid bismuth pool was used as a source of  $Bi^{+3}$ , successful polarograms were obtained with the diffusion current proportional to  $Bi^{+3}$  concentration. The Zn-Bi system has been studied, and the diffusion coefficient of zinc in bismuth was found to be  $5.2 \times 10^{-5}$  cm<sup>2</sup>/sec at 450°C.

(4) The recently developed method of utilizing differences in internal conversion as a means of isotopic analysis was successfully applied to the determination of the degree of burn-up in uranium fuel.

(5) Apparent discrepancies in analyses of  $Te^{132}$ - $I^{132}$  product solution have been found to be the result of hydrolytic precipitation of tellurium at nearly neutral pH during the sampling, dilution, and storage, rather than of the analytical procedures *per se*.

(6) A spectrographic procedure with dc arc excitation is being developed for determining manganese in blood with cobalt used as an internal standard. Radioactive tracers have been used to establish a satisfactory ashing procedure. The approximate concentrations of manganese in dog plasma have been estimated.

(7) Exploratory studies of controlled potential coulometry as a means of determining organic disulfide compounds have been undertaken in connection with a study of insulin being made in the Medical Department.

(8) Optimum conditions have been determined for activation analysis of trace amounts of gold in brain tissue samples.

(9) A more reproducible fluorimetric method for determining uranium has been established. The procedure involves extracting the uranium with an organic ketone from an aqueous phase saturated with  $Al(NO_3)_3$ , evaporating an aliquot of the organic phase, and fusing it with a LiF-NaF flux. A procedure that permits fluorimetric determination of 0.1 to 10 ppm uranium in sodium was also developed.

(10) Spectrophotometric determination of titanium as the cupferronate in Pb-Bi alloy was shown

to be feasible. Bismuth and lead are kept in aqueous phase with EDTA at a pH of 5.5, the cupferronate being extracted with methyl isobutyl ketone and measured at 350 m $\mu$ .

(11) Since separation of Sr<sup>90</sup> from calcium is not complete in the usual method of Sr<sup>90</sup> determination if calcium is present in large amounts, a "flame photometric" technique has been developed for determining the Sr<sup>90</sup> chemical yield.

(12) In the analysis of U-Bi alloy for zirconium, the results should show elemental (not oxidized) zirconium. Present analytical methods were tested and were found to satisfy this requirement.

(13) It was found that the solubility of thorium in a Bi-ThO<sub>2</sub> slurry cannot be determined by a procedure involving dissolution of the bismuth in 50% HNO<sub>3</sub>, inasmuch as some of the "insoluble" ThO<sub>2</sub> is also dissolved.

(14) The effect of dissolved sodium on the spectrographic analysis of bismuth for magnesium and corrosion products was determined.

(15) A satisfactory procedure for analyzing for uranium and zirconium in a bromine-ethyl acetate solution of Zircaloy was developed. Early experiments had indicated the feasibility of determining fluoride in Zircaloy solutions by flame photometry with the Ca-F molecular emission band at 529 m $\mu$ , but more recent work has indicated that the presence of zirconium suppresses this band to unacceptable levels.

### Other Activities

A total of 1303 routine radiographs were made during the year. These included radiographs that showed flaws in an 8½-in. stainless steel billet which was to have been machined into a pressure vessel.

The equipping of the new hot metallurgical cell was, for practical purposes, completed.

Long-lived (2.3-yr half-life) Cs<sup>134</sup> sources were prepared for use by customers as standards for determining the efficiency of counters for I<sup>132</sup>.

During the year a leak was discovered in the calandria of the waste concentration plant. The plant was shut down and the leak was repaired. A new calandria has been purchased and will be installed at a future date. Other losses in operating time were caused by a three-week strike in March and by the short-circuiting of a 60-hp motor which had to be sent off-site for repair.

Early in the fiscal year the volume of "D" waste being received increased to such an extent that the

Table 6  
Liquid Waste Operations

“F” Waste Discharged to the Sewer				
Period	Volume, gal	Average activity level, C/ml		Total activity, mC
Fiscal 1960	4,535,735	$1.72 \times 10^{-11}$		295.4
Fiscal 1959	5,007,315	$9.6 \times 10^{-12}$		183.4
Waste Concentration				
Feed, gal			659,565	
Average volume reduction			128:1	
Drummed concentrate, gal			5,130	
Waste Inventory in Gallons				
	Fiscal 1960		Fiscal 1959	
	“D” waste	“A” waste	“D” waste	“A” waste
Volume on hand at beginning of fiscal year	127,190	4,372	94,020	3,760
Volume received during year	633,395	70	459,630	117
Volume disposed of	656,100 <sup>a</sup>	2,500 <sup>b</sup>	426,460 <sup>a</sup>	—
Volume on hand at end of fiscal year	104,485	1,942	127,190	4,372
<sup>a</sup> To evaporator.				
<sup>b</sup> To Igloo Area.				

<sup>a</sup>To evaporator.

<sup>b</sup>To Igloo Area.

waste concentration plant was put on two-shift operation for several months in order to reduce the "D" waste inventory to a more practical level.

Table 6 indicates the magnitude of the liquid waste operation for the past two years.

### HEALTH PHYSICS

The staff of the Health Physics Division has grown from 47 to 55 during the year, partly because of the continuing growth in the size and complexity of Laboratory operations and partly because of the resumption of operation of the Cosmotron. Service activities such as personnel monitoring, area monitoring, waste disposal, reclamation, hazard evaluation, and bio-assay have continued to be the major responsibilities. The only major facility acquired during the year was a very

sensitive scintillation counter using two 4-in. NaI crystals in a 9-in.-thick steel shield. The counter will be used primarily for counting of fission foils from seven criticality accident dosimeters of the ORNL type that have been installed in suitable locations. However, the counter has several other applications.

In addition to the applied research and developmental work intimately related to the service activities listed above, two special projects were in progress. The first was a study of the particulate activity in the effluent air from the Graphite Research Reactor, carried out during the summers of 1958 and 1959 by an AEC Fellow in Health Physics, which has been accepted by the University of Rochester as a thesis for the degree of Master of Arts. The particulate activity in question was studied by means of microscopic size counts, gamma spectrography, and radioactive decay measurements. The activity, amounting to several curies per day, was found to be contained mostly in particles  $< 1 \mu$  in size and to consist predominantly of iodine isotopes. This project will be continued.

The second research project was a continuation of a study of the effects on rats and monkeys of partial-body exposures to x-rays. This project, carried out in cooperation with the Biology Department, is discussed under the heading, "Studies in Radiation Biology."

The customary program of on-the-job training in applied health physics was provided during the summer for a group of AEC Fellows. Several foreign nationals and U.S. Public Health Service officers were included in the program, the total enrollment being 20, the maximum that can be accommodated. As in the past, short training programs were provided for numerous foreign and domestic visitors.

### Area Monitoring

As of January 1, 1960, monitoring to evaluate the new  $\text{Ar}^{41}$  levels resulting from reloading of the Graphite Reactor had been in progress for 18 mo. A tabulation of data for calendar 1959 showed that values at the edge of the site were  $< 1/10$  the maximum permissible value. During the spring of 1960 the monitoring stations were relocated: four are now on or near the site's perimeter; two at some distance from the site, for reference; and three at new locations, closer to the stack than heretofore, where data on radiation levels are desired. The highest average level at the site bounda-

ry was obtained northeast of the stack and amounted to 0.84 mr/wk. The highest level noted for an individual week was 4 mr/wk.

Routine measurements of the rate of deposition of radioactivity in rainfall and settled dust continued to be made by the pot collection method. However, accumulated weekly samples were analyzed rather than daily samples. The average fallout rate was 32,400 dis/min/m<sup>2</sup>/wk (4630 per day), which is 54% of that noted during the previous year. However, there was a notable peak of 212,800 dis/min/m<sup>2</sup> for the week ending February 29. This one week accounted for 55% of the total fallout during the year. The next highest values were 13,000, 15,700, and 27,300 dis/min/m<sup>2</sup>, for the weeks ending July 20, October 26, and December 14, respectively.

### Personnel Monitoring

The use of personnel monitoring equipment during fiscal 1960 is shown in Table 7, and the exposure record for calendar 1959 is summarized in Tables 8 and 9. All exposures were  $< 3$  rem per 13-wk period except in the case of two individuals

Table 7  
Use of Personnel Monitoring Equipment

	Fiscal 1960	Fiscal 1959
Pairs of pocket chambers	680	812
Regular film badges	735	642
Visitors' film badges	288	263
Total film badges	1023	905
Neutron films used	838	824
Neutron films read	284	291

Figures are weekly averages for each fiscal year.

Table 8  
Exposure Totals and Averages for Calendar 1959

Type of exposure	Total for all individuals	Average per individual	Average per individual per week
$\beta$ (rad)	170	0.086	0.0017
$\gamma$ (r)	271	0.137	0.0026
$n$ (rem)	21	0.011	0.0002
$\gamma+n$	292	0.147	0.0028
E.I.*	377	0.190	0.0037

\*E.I. = Exposure Index ( $1/2$  of  $\beta$  exposure in rad +  $\gamma$  exposure in r + neutron exposure in rem).

Table 9

Distribution of Exposures of Individuals Receiving Regular Personnel Monitoring Service

Exposure interval*	Calendar 1958	Calendar 1959	Integrated**
0.0- 0.4	1415	1815	1425
0.5- 0.9	83	66	140
1.0- 1.4	35	25	68
1.5- 1.9	13	16	54
2.0- 2.4	13	16	37
2.5- 2.9	11	8	27
3.0- 3.4	6	11	32
3.5- 3.9	13	2	23
4.0- 4.9	7	12	35
5.0- 5.9	5	3	23
6.0- 6.9	0	2	16
7.0- 7.9	4	3	9
8.0- 9.9	4	2	21
10.0-11.9	0	0	18
12.0-14.9	0	0	10
15.0-19.9	0	0	8
20.0-24.9	0	0	11
25.0-29.9	0	0	6
30.0-39.9	0	0	14
40.0-49.9	0	0	0
50.0-59.9	0	0	1
60.0-69.9	0	0	2
70.0-79.9	0	0	1
Total	1609	1981	1981

\*Tabulation is in terms of values of exposure index as defined in Table 8.

\*\*Number of persons receiving stated exposure during their total BNL employment period.

at the Graphite Research Reactor who received exposures of 4.9 and 3.9 rem respectively in one 13-wk interval. In no case has an individual exceeded the prescribed limit on integrated exposure. Despite an increase of 23% in the number of individuals monitored, the exposure total for the whole staff decreased by 17%.

### Studies in Radiation Biology

A number of studies on the effects of partial-body irradiation of monkeys and dogs are being continued in cooperation with the Biology Department. These studies may be divided into two general groups, those concerned with the immediate or early effects of irradiation, and those concerned with the delayed or late effects.

In the first category, work just completed has led to characterization of the changes in blood serum proteins following both total-body and partial-body exposures in the LD<sub>50</sub> 30-day range. This work is being carried out in cooperation with Dr. D.G. Baker of the Best Institute at the University of Toronto and is related to the studies of changes in the behavior pattern of irradiated rats mentioned in last year's report.

In the studies on long-term effects, work has been completed on the shortening of life span and the induction of tumors following fractionated doses of x-rays.

Also of particular interest in the study of late effects is the work being done in cooperation with Dr. J.R.M. Innes of the Biology Department. Complete pathological examination of animals showing the aggressive behavior pattern previously described indicates the presence of a progressive demyelinating lesion of the spinal cord in the irradiated area. This lesion is of interest for several reasons: its ultimate result is death, it appears as a late effect (5 to 18 mo postirradiation), and it is produced by doses in the therapeutic range (2500 to 4000 r delivered to a limited area of the spine).

Detailed histochemical techniques are now being used for the purpose of further characterizing the lesion. The effect has been consistently produced in rats, and early results of work with monkeys indicate that it can also be produced in primates. Details of this work were presented at the 1960 meetings of the Radiation Research Society in San Francisco and the International Academy of Pathology in London, England.

### General Safety

Injury frequency rates and accident costs have continued their generally downward trend, as shown in Figure 2. The accidents seldom are directly related to the scientific effort, the majority being falls, back strains, or the result of misuse of hand tools.

Since an analysis of accident investigations indicated that most cases were entirely attributable to negligence on the part of the injured person, greater efforts were directed toward safety promotion. These included a Safety Awareness Issue of *The Bulletin Board* in December 1959, as well as regular features in other issues, and special posters pertinent to BNL safety problems. Safety training was intensified through indoctrination lectures for new employees, seminars for supervisors, and a pilot

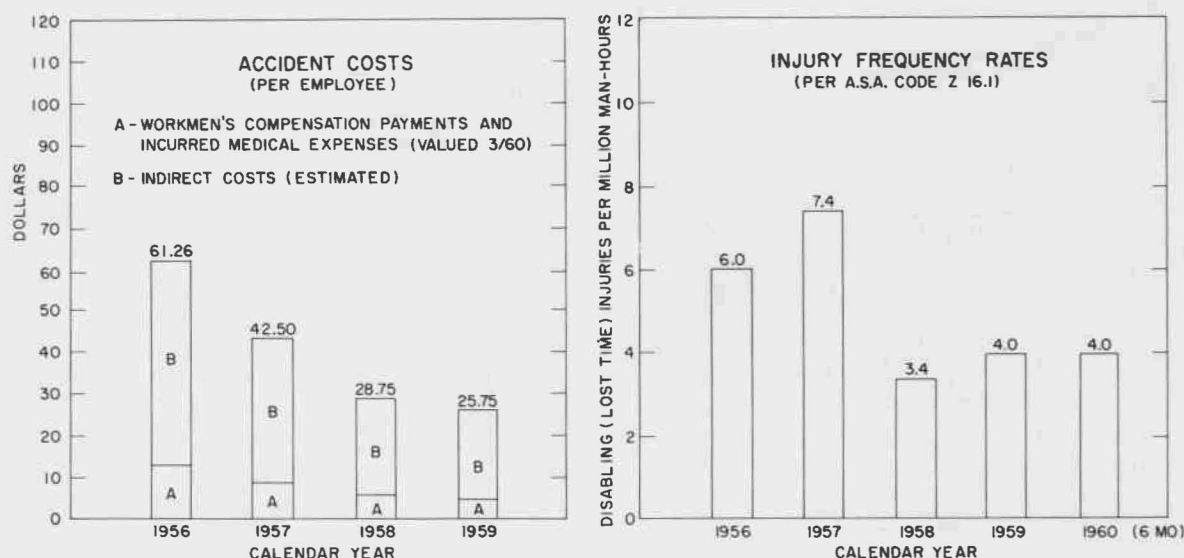


Figure 2.

safety refresher course for foremen in the Maintenance Division. Training in general safety, fire protection engineering, and industrial hygiene was also given to the AEC Health Physics Fellows.

Technical problems continued to receive considerable attention, particularly those involved in planning for the use of liquid hydrogen at the AGS. A paper on liquid hydrogen safety by L.A. Baker, Safety Services Manager, was published in the April 1960 issue of *National Safety News*. Suggestions were made to the AEC Safety Branch concerning research needed in this field, and developmental work was started on a system for extinguishing hydrogen vent stack fires. In connection with the routine review of plans and specifications for all new facilities, standards for fire protection and safety in the design of new buildings were issued for the use of engineers and architects designing BNL facilities. A number of pilot plants were also reviewed, and safety recommendations affecting design and operating procedures were made.

The self-inspection system was extended to all BNL departments, and a schedule was established for calendar 1960. Regular inspection of emergency self-contained (bottled air) breathing apparatus throughout the Laboratory was formally taken over by the Safety Services Office, which also conducts training in the use of this equipment.

### Waste Disposal

A 15-mo accumulation of radioactive wastes packaged in 1352 drums each containing 55 gal was transported to Floyd Bennett Field and

dumped at sea by Navy LST during September 1959. The use of 55-gal drums for this purpose has now been discontinued because of the greatly increased concentrations and amounts of activity and the need to accommodate larger objects. Five types of reinforced concrete vaults, weighing 10 tons when filled, have been developed and are now being used to package radioactive wastes (see Figure 3). After loading, any remaining space is filled with concrete, venting tubes are added when the contents are compressible, and a reinforced concrete top is poured.

The new packaging method has been very successful in handling the increased volume of higher specific activity slurry from the liquid waste evaporation plant. This slurry is pumped into one of the concrete vaults, cement is added simultaneously from a conveyor, and the mixture is stirred mechanically. The procedure is controlled remotely so that exposure during packaging is negligible. The large block of solidified slurry is self-shielding to a considerable extent, and the 6-in. outer layer of clean concrete reduces the surface radiation level to  $\approx 200$  mr/hr. The slurry currently assays at 190 mC/gal. About 20,000 gal/yr are being packaged. One vault will hold an amount of slurry that formerly would have required 50 drums.

The concrete vaults will be stored locally for the immediate future until arrangements for disposal at sea or at a land disposal site can be made. A preliminary cost study has indicated that the vault method will be considerably less expensive than the drum method for either sea or land disposal.



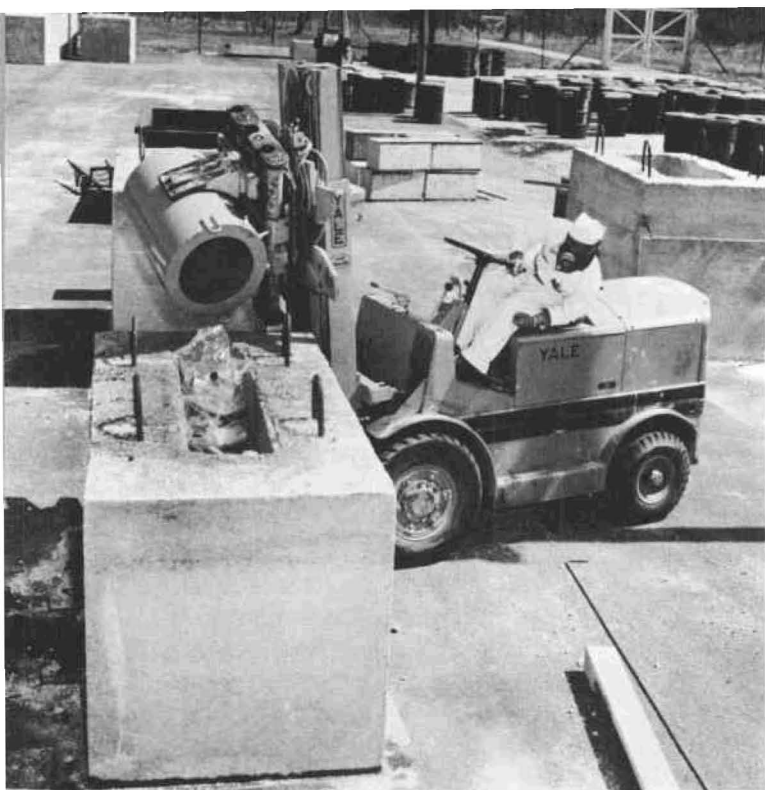


Figure 3. Emptying highly radioactive waste from a lead container into a disposal vault with 17-in.-thick reinforced concrete walls, for ultimate disposal by sea burial.

Data relating to the liquid waste system are presented in Table 10. The amount of activity leaving the site in the stream to which the effluent is discharged totaled 130  $\mu\text{C}$  during fiscal 1960, and the average concentration was  $1.04 \times 10^{-7} \mu\text{C/cc}$ .

### METEOROLOGY

The service activities of the Meteorology Group include routine weather observations and a minimum forecast program. In addition, a number of special studies are conducted each year on request. Currently important is a cooperative effort with the Health Physics Division to re-evaluate the contribution of the Graphite Research Reactor to the local radiation dose rate. This program was necessitated by the significant changes in both the radiation background of the area and the operating characteristics of the reactor since the first thorough study was completed in the early 1950's. By means of a computer program, the hourly meteorological records were used to separate those radiation monitoring stations affected by the reactor from those that were not. The completed survey, which covers a full year's data, shows clearly that the considerable fluctuation in the local background is due to sources unrelated to the Labora-

tory, and that the contribution from the reactor continues to be at barely detectable levels.

Another important item in the service category has been a meteorological evaluation for the High Flux Beam Research Reactor facility. In this study, the complete set of computer programs prepared by the Meteorology Group was utilized to accomplish, in a matter of hours, diffusion and deposition estimates that would have been virtually unobtainable a few years ago.

The extension of the automatic data collection system to include the recording of climatological data has been a welcome addition to the service capabilities of the Group. In March 1960, the recording of all necessary wind, temperature, solar radiation, and precipitation data began on a routine basis, with the collection system scanning each element at 6-min intervals, 24 hr per day. This information is recorded on paper tape, converted to cards, and then processed to provide a printed record of 6-min instantaneous data, hourly and daily averages, and finally, monthly summaries. In due course, this same process will be accomplished by the Merlin computer directly from the tape, and the IBM card conversion will be eliminated.

Table 10

Summary of Liquid Waste Data

	Fiscal 1960	Fiscal 1959
Input to filter beds, gal/day	523,300	514,800
Output from filter beds, gal/day	464,700	395,200
Net loss in filter beds, %	11.2	23.2
Stream above discharge point, gal/day	604,600	1,835,000
Stream at site boundary, gal/day	931,400	2,208,000
Rainfall, in./mo.	3.2	3.7
Activity concentration at input to filter beds, C/cc	$7.3 \times 10^{-13}$	$5.7 \times 10^{-13}$
Activity concentration at output from filter beds, C/cc	$2.7 \times 10^{-13}$	$2.1 \times 10^{-13}$
Reduction in activity concentration, %	63	63
Activity at input to filter beds, mC/mo	42.4	33.0
Activity at output from filter beds, mC/mo	15.3	8.9
Percent of permissible	28	26

Figures are averages for each fiscal year.

## APPLIED MATHEMATICS COMPUTATIONAL SERVICES

As in past years, the Applied Mathematics Division supplied mathematical services for the Laboratory's scientific staff. These included general aid in programming and mathematical analysis, the full development of the Merlin computer, and the operation of the IBM 704 computer for the Laboratory at the New York University-Atomic Energy Commission Computing Facility.

The developmental work of the Merlin programming and assembly system has been described in the Applied Mathematics section of this report. Two lecture series on Merlin were presented to the Laboratory staff during the year. The first was addressed to those members of the Laboratory who had extensive knowledge of computer usage, while the second was directed toward personnel with limited computer experience. Two internal reports describing Merlin and its programming system were prepared to supplement the lecture series. A training program for beginners is being planned for the coming year.

Laboratory requirements for time on the IBM 704 computer increased steadily throughout the year. By the end of the fiscal year, the machine was being used  $\approx 1$  shift/day, 5 days/wk. Usage for the year totaled 1214 hr, as compared to 376 hr in fiscal 1959. The use of computers as scientific tools in research is expected to increase at a rapid pace, with requirements in fiscal 1961 approximating 2800 hr and continuing to increase in future years. These increases are attributable in part to the growing awareness of the Laboratory staff of new applications of computer techniques. Another factor is the need for analysis of bubble

Table 11

Distribution of IBM 704 Time by BNL Departments  
During Fiscal 1960

Department	Usage, hr	Usage, %
Accelerator Development	46.24	3.8
Applied Mathematics Division	52.46	4.3
Chemistry	54.60	4.5
Cosmotron	57.28	4.7
Instrumentation and Health Physics	46.61	3.9
Medical	14.83	1.2
Nuclear Engineering	370.34	30.5
Physics	572.32	47.1
Total	1,214.68	100.0

Table 12

Distribution of IBM 610 Time by BNL Departments  
During Fiscal 1960

Department	Usage, hr	Usage, %
Accelerator Development	19.00	1.4
Applied Mathematics Division	155.75	11.2
Chemistry	136.25	9.8
Cosmotron	66.75	4.8
Medical	118.75	8.5
Nuclear Engineering	470.25	33.7
Physics	425.50	30.6
Total	1,392.25	100.0

chamber data in connection with the high energy physics program. It is clear that in this latter field computer analysis of data will become increasingly important.

Table 11 shows the distribution of IBM 704 time among the various scientific departments. Usage of this computer by the Nuclear Engineering and Physics Departments continues to increase, and in fiscal 1960 accounted for 77.6% of the total. The Medical Department made use of the IBM 704 computer for the first time this year.

Since the Laboratory's use of computers at the NYU Facility is limited by the time that can be made available, and since this situation is expected to become more critical with increasing BNL computer requirements, initial plans have been made for the acquisition by BNL of another large digital computer.

The use of commercial courier service to transport computational materials to and from the NYU Facility was continued. A second IBM 704 computer operator has been engaged, and plans for a considerable increase in the programming staff were initiated.

The IBM 610 computer, which is a simple, small-scale machine installed at BNL in 1958 to assist in the processing of large desk calculator problems, was placed on a self-service basis and supported with a programming consultation service. Distribution of 610 usage is shown in Table 12.

## MECHANICAL ENGINEERING

To meet the demand of various departments of the Laboratory for greater engineering support, the Mechanical Engineering Division has added 8

engineers and 7 designers during the year, most of whom have been assigned to the Bubble Chamber Group of the Physics Department and the Experimental Support Group at the Cosmotron (see Table 13).

The members of the Division assigned to the Accelerator Development Department have completed the design, fabrication, and assembly of mechanical parts of the Alternating Gradient Synchrotron (AGS) and are making minor modifications. The design of circular quadrupole magnets, rectangular quadrupole magnets, and bending magnets has been completed. Most of the 43 magnets, which will be used in experimental beams, have been ordered and some have been fabricated. Targeting equipment and special straight sections have been designed. A modular beam separator with a unit length of 16 ft and an aperture 4 in. high by 24 in. wide is being designed. It will be housed in a vacuum tank that is part of the magnetic circuit and is so arranged that units can be coupled together to form separators of different lengths. Fabrication of six of these units is planned.

Specifications and drawings have been completed for modification of the AGS Target Building to accommodate bubble chambers using explosive gases, and studies have been made of the layout of mechanical equipment and services in the new experimental area.

Division personnel assigned to the Nuclear Engineering Department have also performed work for the Reactor Division. The work on bismuth and salt loops decreased with the termination of the LMFR project, but new programs for mercury and NaK heat transfer research required pumping loops.

Work for the Reactor Physics Experimental Group continued at an increasing rate to require vessels, fuel element assemblies, control rod drives, and handling machinery for critical assemblies. Work was done on equipment for encapsulating, welding, testing, handling, arranging in proper configurations, and storing  $\text{Co}^{60}$  sources for a radiation source development laboratory, which also necessitated liaison with architectural engineering firms.

Equipment was designed for high temperature pyrographite research, arc image furnaces, and fuel element testing. Designs and estimates for evaluation of  $\text{UO}_2$ -sodium reactor concepts were completed for the Reactor Evaluation Group.

Table 13

Assignment of Mechanical Engineering Division  
Personnel as of June 30, 1960

Department	Engi- neers	Designers and Draftsmen	Secretarial and Clerical
Accelerator Development	11½	19	½
Central Design Group	½	6	1½
Cosmotron	11	6	0
Nuclear Engineering			
General design	3	14	1
High flux reactor	2	5	0
Physics			
General design	0	5	0
Bubble chamber	13	8	0
Total	41	63	3

The possibility of locating new vertical holes in the Graphite Research Reactor was examined, and studies were made of chemonuclear loops for these holes. Hydraulic studies were made in connection with plans to increase the power level of the Medical Research Reactor.

The specifications of new cooling fans for the Graphite Research Reactor and engineering liaison for their installation were completed. The fans are performing satisfactorily, and the expected power economy is being realized.

Division personnel are assigned to key positions in connection with the construction of the High Flux Beam Research Reactor and are responsible for assisting in conceptual design and checking of architectural engineering work in the following areas: general layout of the building and services, layout of reactor vessel control rods, and the overall cooling system. Also included are core shroud design, electrical systems layout, general piping and instrument diagrams, stress analyses, thermal and biological shield design, experimental facilities design, and fuel handling.

Personnel of the Mechanical Engineering Division assigned to the Cosmotron were concerned with its maintenance and improvement. A second Piccioni beam ejection magnet and focusing shims were constructed and installed for extraction of Beam 3, and a new series inductor ripple filter was completed. A study has been made for redesign of the magnet cooling water system to allow conversion from cooling towers to the use of well water and a heat exchanger. This will provide greater cooling capacity for a higher repetition rate and

for flat-topping the magnetic field at the end of the cycle. In addition, a 10-ton gantry crane, fashioned from the overhead hoists formerly used in the Cosmotron, was installed in the Assembly Building.

The design and fabrication of experimental equipment constituted a major effort. Design was completed of a liquid deuterium target and deuterium recovery system, and specifications and drawings were made for a 10-ft-long liquid hydrogen target for the AGS. The first of two 15-ft-long beam separators was completed and placed in operation, and the second is almost finished. A high-pressure Cerenkov counter was designed and manufactured as well as a secondary emission ionization chamber.

Division personnel have established regular procedures for assisting experimental physics groups in setting up and carrying out complex experiments on the Cosmotron floor.

Members of the Division continued to operate and improve the hydrogen liquefier facility. A new gas purification system was added, and the compressor was speeded up to increase the output to 90 liters of liquid hydrogen per hr. This year 40,000 liters of liquid hydrogen were produced.

Personnel assigned to the Central Design Group performed work and supervised installation of equipment for many Laboratory departments. Among the projects were the design and installation of three neutron spectrometers, the design of a liquid nitrogen transfer line at the Graphite Research Reactor, a whole-animal grinder for the Biology Department, an isokinetic sampling device for the Meteorology Group, a pressure homogenizer for the Medical Department, and a coordinatometer for the Physics Department.

As already indicated, one of the major assignments of the Division is the design of the 80-in. liquid hydrogen bubble chamber. Assistance is also given in operating the other bubble chambers for which the Bubble Chamber Group of the Physics Department is responsible.

A computer analysis has been made of the expansion system of the 80-in. bubble chamber, and based on these results the design of the piston expander is proceeding. Detailed stress analyses of the expansion system, vacuum chamber, magnet coils, magnet iron, and magnet base have been performed. In addition, detailed mechanical design has been started on many of the above components.

The design and selection of components for the Freon, liquid nitrogen, and liquid hydrogen refrigeration systems has been completed, and bids are being obtained on the compressors for these systems. Division personnel have been closely connected with the design of the building and services for the bubble chamber.

Another group of Division personnel has been performing general design work for the Physics Department, including neutron spectrometer equipment and scanning equipment for the Bubble Chamber Group. In addition, work has been done on the design and fabrication of a new set of dees for the 60-in. cyclotron.

### MACHINE SHOPS

The Machine Shops, while carrying a substantial workload of small and light work, have been increasingly engaged in the fabrication of large, heavy units, and in work requiring specialized equipment. There have been increases in small-quantity production work, larger and heavier weldments, and lighter and more critical welded assemblies, coupled with a general demand for high quality welding. Much more frequently than in the past the Machine Shops are being called upon to make complete units, including fabrication of component parts, assembly, checking of operation, and delivery.

Total productive hours for fiscal 1960 show an increase of  $<1\%$  over fiscal 1959. Additional personnel in 1961 should provide an increase of 10 to 12% in productive hours. The division of the workload has remained quite constant, with approximately three fourths being machining, and the remainder sheet metal and welding work. In the latter part of the year the Machine Maintenance Section was transferred from the Plant Maintenance Division to this Division.

The procurement in fiscal 1960 of replacements and new equipment has further increased the capability of the Division and has brought the equipment inventory closer to a proper balance. New welding equipment and heavy machine tools were the areas of maximum emphasis.

The Standards Section moved to new air-conditioned quarters in the latter half of the year. The more efficient layout, increased space, and excellent lighting have all contributed to easier and faster inspection and maintenance of a higher degree of accuracy. The workload has continued

to increase, and additional inspection personnel will probably be required to meet future demands. The Technical Consulting Group provided advice and technical assistance at a somewhat lower level than in the previous year, with the Nuclear Engineering, Cosmotron, Chemistry, and Medical Departments accounting for more than two thirds of the projects undertaken.

The contributions of the several departments to the workload of the Machine Shops are indicated in Table 14. The most significant change was the almost doubled workload contributed by the Accelerator Development Department. The Nuclear Engineering, Physics, and Cosmotron Departments continued to account for over two thirds of the total workload.

More than 1500 projects were processed through intralaboratory requisitions during fiscal 1960, exclusive of numerous small jobs undertaken on a short-order basis. Only a few of the larger or more unusual projects, exemplifying the type of work done and the wide variety of problems encountered, can be mentioned here: Fabrication of a stainless steel liquid nitrogen Dewar, 20 in. in diameter and 10 ft long; construction of the H  $6 \times 12$ -in. Mk2 bending magnet for the Cosmotron; fabrication of four complete control rod mechanisms; construction and installation of a rotating shutter mechanism for the spectrometer in Experimental Hole W-34; machining of the linac buncher assembly; modification of a gantry crane with 54-ft girders for the Cosmotron; fabrication of a fuel handling mock-up platform as-

sembly 33 ft high; development of stainless steel bone needles for the Medical Department; building of two digitizers for the Cosmotron; construction of the inflection plate alignment fixture for the AGS; machining of enriched  $\text{UO}_2$  fuel elements; and machining of a pure tantalum ion source for the Chemistry Department. Also included were machining of a Pu-U alloy in a dry box specially designed and manufactured in this Division; welding of 0.005-in. stainless steel windows for the liquid nitrogen Dewar; welding of an 0.008-in. deuterium target for the Cosmotron; the close tolerance welding of 0.007-in. stainless steel sheets to stainless steel plates 3 in. thick by 18 ft long; and fabrication of a BNL fast chopper rotor for the Phillips Petroleum Company.

### TECHNICAL INFORMATION

The construction of an extension to the Research Library Building, consisting of 3600 square feet at the west end, was completed during the past winter. This addition now accommodates ten double-faced ranges of steel shelving and 85 file cabinets. The resultant alterations in the original part of the building have provided increased space in the periodical room and the lobby. The relocation of the Library's files of 35 abstract journals in the new extension has proved to be of considerable convenience to both the Laboratory's scientists and the Library's staff.

Activity in the Circulation and Reference Sections of the Library has increased markedly. In the former, as the collection has grown, its use has also. The latter reports the receipt of a greater number of reports than in previous years; part of this increase reflects the release of documents originally published on a classified basis. The number of requests for information and for the preparation of bibliographies continues to mount.

During the year 679 reports, of which 343 are classified as secret and 336 as confidential, were received in the Classified Library, although only one classified report was issued by BNL. In this same period, 676 reports were declassified and 384 report copies were destroyed, upon receipt of the necessary official notification from the AEC. The existing classified holdings, as disclosed by the annual inventory, number 9200 secret and 6564 confidential documents. Inventory of the correspondence holdings showed a total of  $\approx 7100$  classified memoranda and other communications in the files.

Table 14

#### Departmental Contributions to Machine Shops Workload

	% Man-hours		
	Fiscal 1959	Fiscal 1960	% Change in workload
Accelerator Development	8.7	15.3	+75.9
Biology	1.6	1.7	+6.2
Chemistry	2.0	2.0	—
Cosmotron	19.5	21.0	+7.7
Instrumentation and			
Health Physics	3.3	1.2	-63.6
Medical	5.4	3.2	-40.7
Nuclear Engineering	35.9	36.6	+1.9
Physics	12.1	11.2	-7.4
Reactor	8.9	6.3	-29.2
Miscellaneous	2.6	1.5	-42.3

The Editorial Group, which provides technical editing services for manuscripts planned for publication as BNL reports, has continued to operate at about the same level of activity as in the past few years. In addition to its editing and proof-reading functions, this group also checks the accompanying artwork and captions and otherwise processes the raw material intended for publication.

A further 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. As an index to the Laboratory's increasing output of such papers, the Information Division processed a total of 607 items (including published abstracts of speeches) last year, as compared with 486 for fiscal 1959, 473 for fiscal 1958, and 401 for fiscal 1957.

The distribution lists for the Laboratory's unclassified reports also continue to grow. Standard distribution is made as prescribed by the AEC, but in addition all bona fide requests from within the national atomic energy project and from cooperating government agencies and universities are honored. With the development of atomic energy programs in many countries, the Division has been able to establish with foreign scientific institutions a number of mutually beneficial exchange arrangements whereby BNL reports are offered in return for foreign journals and reports. Three important journals in this category, not generally available in the United States but now being received in the BNL Library, are *Sinica Scientia*, *Science Abstracts of China*, and the *Chinese Medical Journal*. Such arrangements can be expected to increase steadily, as research in the nuclear sciences extends to all parts of the world and increases in scope.

#### PHOTOGRAPHY AND GRAPHIC ARTS

The activities of the Photography Group are indicated in Table 15. These figures, however, do not include a number of photographic commitments throughout the Laboratory involving the Group's cooperation.

The increase in the number of bubble chambers at the Cosmotron and the experimental program planned for the AGS will require the photographing of several million bubble chamber events per year. For this purpose a continuous film processing

Table 15

	Fiscal year		
	1960	1959	1958
Photographs	9,133	7,135	7,198
Photomicrographs	1,898	1,405	1,732
Lantern slides	7,576	8,138	5,946
Prints	46,747	47,533	57,255
Photostats, Xerox	32,698	20,560	22,127
Ozalid prints	502,227	430,193	429,625
Film processed, ft	196,532	1,174	86,592
Charts and graphs drawn	3,058	3,091	2,615

Table 16

	Fiscal year		
	1960	1959	1958
Offset pages	9,921,637	8,641,645	6,361,665
Offset negatives	9,050	9,482	8,738
Mimeo impressions	1,560,263	1,567,489	1,228,386
Stencils cut	953	814	728
Stencils run	11,530	12,410	11,424
Sheets collated and bound	2,996,192	3,679,450	2,388,700
Report copies issued	115,473	109,780	65,715

machine has been designed and installed, and is now operating satisfactorily at its capacity of 46 ft of film per min. Two people have been added to the staff to operate the machine and perform control sensitometry.

The nuclear emulsion laboratory has done development work for several cooperating institutions, and has devoted considerable effort to an investigation of the processing technique in relation to spurious scattering and distortion.

Two short motion pictures were completed, one on the fabrication of the linac drift tubes and another on the assembly of the ferrite accelerating cavities of the AGS.

The Laboratory has received approval from the Joint Congressional Committee on Printing for the rental of a second Fotosetter from the Intertype Corporation. This will double the Graphic Arts Group's composing capacity, the most heavily burdened area of graphic reproduction in the past. Plans have been approved for the installation of a system to control the humidity and temperature in the printing plant; this will add greatly to the efficiency of the paper-handling machinery.

Reproduction figures for the past three years are given in Table 16.

# 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 and Students

The numbers of scientists and students holding appointments at Brookhaven on May 31, 1959 and 1960, are shown in Table 1. Totals for the regular staff seemingly belie the continued, gradual increase in Laboratory personnel. This is due to a change in the method of reporting Junior Scientists. Since May 1959, individuals in this category have not been included in statistics relating exclusively to the scientific staff. This accounts, in part, for the proportionate increase in

the number of staff members with doctoral degrees.

Turnover continued at a significant rate. During the year, 41 scientists were appointed to the regular staff, 36 terminated, and 7 were transferred to other categories.

The number of salaried visitors, as of May 31, 1960, was approximately the same as last year. During each of the four previous years, this number increased by more than 10%. Noteworthy among salaried visitors are Research Associates, whose participation has become an increasingly important fraction of the total research effort at Brookhaven. Appointments as Research Associates are similar to postdoctoral fellowships and are limited in term. Consequently, turnover is particularly high. During the year, 46 scientists began appointments as Research Associates, and 38

Table 1  
Scientific Staff and Students on May 31, 1959 and 1960

	Visitors					
	Regular staff		Salaried		Nonsalaried	
	1960	1959	1960	1959	1960	1959
By appointment category, Staff						
Senior Scientist	48	46	0	1	11	13
Scientist	95	87	5	2	104	78
Associate Scientist	103	103	6	3	86	77
Assistant Scientist	63	59	1	4	72	60
Research Associate	—	—	67	62	17	14
Junior Scientist*	—	16	—	2	—	5
By appointment category, Students						
Junior Research Associate	—	—	2	6	40	28
Research Assistant	—	—	0	0	17	12
Total	309	311	81	80	347**	287†
By academic degree						
Ph.D. or M.D.	187	175	76	68	236	192
Master	46	42	5	5	56	41
Bachelor	72	91	0	7	51	47
No degree	4	3	0	0	4	7

\*Effective May 1959, Junior Scientist became a nonscientific staff category.

\*\*57 of these appointees were at BNL on a full-time basis.

†66 of these appointees were at BNL on a full-time basis.



Table 2

Classification of Visiting Scientists and Students Participating in BNL Program  
for Period of One Month or More, June 1, 1959 - May 31, 1960

	Guests and salaried visitors				1959 Summer program		Total	
	More than 3 months		Less than 3 months		Salaried	Guest	Individuals	Institutions
	Salaried	Guest	Salaried	Guest				
University staff	18	22	4	22	57	37	160	74
Thesis students	5	18	0	9	1	11	44	15
Student Research Assistants	0	1	2	6	97	36	142	62
Subtotal	23	41	6	37	155	84	346	108 different
Industry	1	20	1	5	0	1	28	14
Other institutions	0	42	3	21	10	9	85	54
Total	24	103	10	63	165	94	459	176 different

others completed their terms. Of the 84 salaried and nonsalaried individuals in this category on May 31, 33 were foreign scientists.

For more than a decade the number of non-salaried visitors holding appointments to the staff has increased each year, and the past year was no exception. The number of such individuals participating at the Laboratory on a full-time basis, however, decreased to the level of two years ago.

May 31, instead of June 30, was selected as the appropriate date for Tables 1 and 2. This was done to exclude from Table 1 statistics relating to individuals at Brookhaven only during the summer, and to include in Table 2 participants in only one summer program.

Table 2 shows the number of visiting scientists and students who worked at the Laboratory for a cumulative period of one month or more between June 1, 1959, and May 31, 1960. Included are 346 faculty members and students from 108 colleges and universities, compared with 259 such individuals from 92 institutions last year, and 358 from 116 institutions two years ago. The total number of visitors here for one month or more increased from 376 last year to 459 this year. During the year, 15 graduate students who had engaged in doctoral investigations completed the research required for their theses.

Appointed visitors at Brookhaven for a cumulative time of less than one month are not included in Table 2. During each of the past two years, 137 scientists and students worked at the Laboratory for periods ranging from one day to one month.

Participation by foreign scientists and students continued to increase. During the fiscal year ending June 30, 1960, 166 individuals from 33 foreign countries worked at Brookhaven. Japan and Great Britain again had the greatest representation, with 23 and 21 individuals, respectively, participating. During the previous year, 145 foreign nationals engaged in research here.

Table 3 contains statistics pertaining to consultants. The use of these specialists, as measured in man-days of service, increased for the second successive year, but is still considerably below the levels of the past decade.

#### Summer Program for 1960

Arrangements have been made for 254 visiting scientists and students to participate at the Laboratory during the summer of 1960. Of this number, 104 are faculty members from 58 educational institutions, and 26 are scientists and engineers from 18 other institutions and organizations.

Table 3

Consultants' Services

	Fiscal year		
	1960	1959	1958
Total contracts in effect June 30	80	80	93
No. of consultants used	36	32	46
No. of man-days of service	336	331	308

In December, announcements of the ninth annual summer student program were sent to 170 colleges and universities. Applications were received from 101 graduate and 483 undergraduate students from 149 schools. Of 105 appointments offered, 75 have been accepted by 27 graduate and 48 undergraduate students representing 46 institutions.

Nineteen Health Physics Fellows, most of whom have completed a graduate curriculum at the University of Rochester, are here for a 10-week training period. In addition, 3 Junior Research Associates plan to work on their doctoral theses at Brookhaven this summer, and 27 other students will work with staff and visiting faculty members as Research Assistants.

### Personnel Management

**Employment.** During the past fiscal year the net increase in the number of employees on the payroll was 125, slightly below last year's increase but a little above the average for the past five or six years. As of June 30 the total staff, including guests and temporary employees, was 2477. Although the demand for highly skilled, specialized technical and professional employees still exceeds the supply, the Laboratory continues to attract such employees with minimum advertising expense, because of its established reputation. Table 4 contains the staff analysis.

**Labor Relations.** New one-year labor contracts were negotiated with the three unions that represent  $\approx 467$  wage employees. A three-week strike in March by 22 of the 24 members of the Oil, Chemical, and Atomic Workers Union was the Laboratory's first experience of a work stoppage. Reactor operations continued on a five-day schedule through the efforts of supervisory personnel, and the Hot Laboratory and Medical Research Reactor facilities were unaffected. Despite the work stoppage, all three unions accepted wage increases approximately equal to those granted nonbargaining-unit employees and comparable to the pattern being followed throughout the nation. Benefit plan modifications incorporated in the labor contracts were identical to those granted all other Laboratory employees.

**Employee Benefit Plans.** Two significant changes were made in the Laboratory group insurance plans. The flat per diem allowance for hospital room and board charges was replaced by an allowance corresponding to the actual charges,

up to and including semiprivate room accommodations. This change eliminated inequities to employees arising from the wide range in the rates charged by various hospitals for similar accommodations. It also permits the plan to keep pace with economic changes without necessitating frequent revision of the per diem allowance. The cost to employees for basic life insurance was eliminated, and a 30% reduction in the cost to employees for supplemental life insurance was put into effect.

The Laboratory's salary and wage policies and benefit program are at least the equivalent of those offered to employees by other research organizations, including Government and educational institutions, and by Atomic Energy Commission contractors. Nevertheless, it is necessary to familiarize the employees, especially the supervisory staff, with the merits of these policies. For this purpose articles are printed in the employees' weekly newspaper, *The Bulletin Board*, seminars for super-

Table 4  
Employment Statistics\*

	Fiscal 1960		Fiscal 1959	
Scientific staff	385		382	
Nonscientific staff	1740		1618	
Total	2125		2000	

Turnover data	Number	Annual rate (%)	Number	Annual Rate (%)
<u>Accessions</u>				
Scientific staff	88	23	93	25
Nonscientific staff	261	16	286	19
Total	349	17	379	20
<u>Separations</u>				
Scientific staff	85	22	81	22
Nonscientific staff	139	9	126	8
Total	224	11	207	11
<u>Net Accessions</u>				
Scientific staff	3	0.8	12	3.2
Nonscientific staff	122	7.5	160	11.0
Total	125	6.2	172	9.4

\*Figures do not include 54 temporary summer nonstudent employees. Guests and temporary student employees are included in Table 2.

visors are conducted, training courses in the problems of supervision are held, and meetings of small groups of supervisors and employees are arranged, as the need arises.

### SECURITY AND PLANT PROTECTION

The Security and Plant Protection Division is a security and service organization in which the security function is of primary concern. Despite the fact that very little classified work is performed at the Laboratory, classified documents, materials, meetings, and visits require the application of security procedures. At the end of the fiscal year 948 staff members held AEC "Q" clearance and 299 held AEC "L" clearance. These staff members required access to classified information or visited restricted installations elsewhere, and such activities involve the services of the Security Office. To this has been added the detailed work required by the additional security procedures imposed by the Department of Defense on the Laboratory and AUI, since both facilities are now cleared for security by that Department. The staff of the Security Office was decreased in size by the elimination of one administrative post.

During the year the Division completed a protection survey of the National Radio Astronomy Observatory at Green Bank, West Virginia, operated by AUI, and undertook to arrange with the Department of Defense for facility clearance so that its research efforts in cooperation with the Armed Services could continue in certain classified areas.

The service function, involving the provision of service and protection to the staff, visitors, and the physical plant, has expanded as the Laboratory has grown in size of staff and number of buildings. The Police Group, through constant rescheduling and with the cooperation of all departments, was able to keep pace with the growth and to provide adequate service and protection without an increase in personnel.

The increased scope of the Division's activities during fiscal 1960 can in part be measured through a comparison of significant services performed. Table 5 presents a three-year comparison of the number of admissions of nonstaff personnel to the site. Each admission required the services of the Division in some way.

The Fire Group continued its fine record in meeting potentially dangerous situations at the

Laboratory. No major fires occurred during the year, and there was a notable drop in the number of responses as compared to fiscal 1959. However, activities in other areas, such as prevention work and fireguard details, increased to such an extent that the limit of the Group's capacity to carry out these necessary and vital duties was reached. The growth of the Laboratory, with the concomitant hazards, places a steadily increasing burden of responsibility on the Fire Group, particularly with respect to its inspection program.

	1960	1959	1958
Responses on site	103	126	113
Responses off site	5	4	2
Investigation - no response	39	31	14
Total	147	161	129

### ARCHITECTURAL PLANNING

During the past year, the diverse activities of the Architectural Planning Division have continued at an accelerated pace and in greater volume.

Several major building projects for which the Brookhaven Area Manager is administering the architect-engineer and construction contracts have required the services of the Division. In the case of the Nuclear Engineering and Physics Buildings, now under construction, review of the shop drawings is being provided on a continuing basis to insure proper interpretation by the architect-engineer and the builders of the requirements of the scientific departments involved.

Three other building projects (an extension to the steam plant, the Volatility Studies Laboratory, and the High Intensity Radiation Development

Table 5

#### Admissions to the Site

	Fiscal year		
	1960	1959	1958
Regular visitors*	46,688	36,472	30,555
Conference visitors	2,593	2,438	4,687
Alien visitors	1,990	2,026	1,931
Subcontractors' employees	21,657	22,576	21,273
Total	72,928	63,512	58,446

\*Includes 15,288 visitors during the Laboratory's annual Visitors' Days.

Laboratory) with a total cost of \$2,300,000 will be under construction by fall of 1960. The necessary reports, working drawings, specifications, and contract documents have been prepared by architect-engineer organizations under contract with the Atomic Energy Commission, and this Division has provided the necessary liaison between BNL, the Brookhaven Area Manager, and the architect-engineer. The problems presented by the High Intensity Radiation Development Laboratory have been both interesting and challenging. This project was divided into two sections, with BNL assuming responsibility for the design, procurement, and installation of the special equipment to be used in the experimental cells, while the AEC retained responsibility for the administration of the architect-engineer and construction contracts for the building and the cells. Inasmuch as this equipment requires precise fitting in the building, it has been essential to provide for the complex coordination of equipment and building parts and for constant liaison between the several agencies involved (the Nuclear Engineering Department, the engineering assistance firm, the architect-engineer, and the Brookhaven Area Office of the AEC) in the management and execution of the project.

In the last quarter of the year, the Division's services were enlisted in connection with several new projects planned for fiscal 1961, including site utilities, an extension to the Metallurgy Building, a critical assembly facility, and quarters for visiting scientists. Authority for the construction of these projects has been granted by an Act of Congress, and in anticipation of the appropriation of the necessary funds (>\$3,000,000) the AEC engaged architectural and engineering organizations to design the buildings. The Division took an active part in the preparation and development of the technical and planning information required by these organizations.

The Division also assisted in the architectural engineering phases of the High Flux Beam Research Reactor. Several studies were made involving building plans, site locations, and comparative cost estimates, and one of the building schemes submitted has been adopted. Similar services will be rendered as the detailed planning proceeds.

One of the responsibilities of the Division was the preparation of the major portion (that relating to buildings) of the facilities section of the budget request for fiscal 1962. This involved preliminary

building plans, site locations, cost estimates, and related descriptive brochures for seven major new buildings or building additions with a total value of almost \$19,000,000, including a laboratory building for the Chemistry Department, a general engineering and services center, an addition to the Hot Laboratory and modifications to the existing structure, a new laboratory wing for the Cosmotron, two facilities for the Biology Department (one for radiobotany and one for pathogen-free animals), and an addition to the Applied Mathematics Center.

During the year, many projects with a total value in excess of \$940,000 were initiated or undertaken; in every case, the work required the preparation of individual proposals, and in all but a few instances the Division furnished the working drawings and specifications, contract administration, and field supervision of the required construction. The principal projects under this heading included

Graphite Research Reactor projects, e.g., remote controls for reactor fans, extension of reactor balconies, canal house crane modifications, and additional power for laboratories	\$ 52,000
Cosmotron projects, including improvements in the magnet cooling water system, transformer substation addition, installation of experimental power supplies (dc), air intake modifications in the motor generator room, and hydrogen gas venting ducts	168,000
Medical Research Center projects, such as an additional heat exchanger for the Medical Research Reactor, reactor air lock improvements, air tempering and summer cooling for the laboratory section (well, cooling coils, discharge basin), ventilation and air conditioning for offices, air conditioning for the whole-body counter, and miscellaneous modifications	305,000
Additions and improvements to the research facilities of the Chemistry and Biology Departments, such as additional chemistry laboratories and offices, air conditioning improvements, a genetics laboratory, and a plant room for growth under controlled conditions	155,000
Ventilating and air conditioning improvements and installation in several research and service areas, such as reactor physics laboratories, the 18-in. cyclotron vault, printing shop, and radioactive materials shop	50,000
Improvements to electrical power system, including an extension to the switch house at the main substation and several modifications in the underground and overhead power lines	88,000

Table 6  
Manpower Utilization

Type of work	Fiscal 1960		Fiscal 1959	
	Productive man-years	Percent of total work	Productive man-years	Percent of total work
Plant utilities operations	47	18	56	22
Building janitor service	38	15	36	14
Decontamination and hot laundry	12	5	11	4
Total plant operations	97	38	103	40
Communications service	16	7	10	4
Transportation, housing, and staff services	18	7	21	8
Total staff services	34	14	31	12
Special services for others	45	18	54	21
Facility improvements	5	2	3	1
Total special services	50	20	57	22
General repairs and maintenance	62	24	43	18
Special long-term maintenance	11	4	21	8
Total maintenance	73	28	64	26

Table 7  
Costs of Supplies, Materials, and Purchased Labor

	Fiscal 1960	Fiscal 1959
Plant utilities operations		
Fuel oil	\$ 235,670	\$ 245,267
Electricity	868,431	728,766
General supplies	38,104	26,360
Building janitor supplies	35,157	23,796
Decontamination and hot laundry supplies	22,624	16,373
Total plant operations	1,199,986	1,040,562
Staff services		
Telephone, teletype, and mail	227,247	184,892
Transportation (gasoline)	20,651	18,900
Housing and cafeteria supplies	77,637	41,691
Total staff services	325,535	245,483
Maintenance		
General materials and purchased labor	202,285	126,766
Special materials and purchased labor	481,000	309,945
Total maintenance	683,285	436,711
Grand total	\$2,208,806	\$1,722,756

Building modifications and additions, including a shop for electronics, an irradiation facility for Solid State Physics, a radio-analytical chemistry laboratory in the Hot Laboratory, a storage building for contaminated materials and equipment, and offices for the Purchasing Division 125,000

### PLANT MAINTENANCE

The service activities of the Plant Maintenance Division include the operation of all Laboratory utilities; the maintenance of all buildings, roads and grounds; and provision of communication services and housing and travel accommodations for all segments of the Laboratory. In addition, skilled rigging, moving, and model-making services are provided in connection with scientific projects. Maintenance of the automotive fleet and heavy mobile equipment is also assigned to Plant Maintenance.

A comparative analysis of the utilization of manpower within the Plant Maintenance Division in fiscal 1959 and 1960 is shown in Table 6, and a comparison of costs incurred in the operation of the Division is presented in Table 7.

Some of the major projects completed by the Plant Maintenance Division during fiscal 1960 are listed below.

- Relocation of a building for use as a meteorology workshop.
- Conversion of a building to serve as archives.
- Relocation of a building for the High Flux Beam Research Reactor project.
- Renovation of the banking facilities on site.
- Relocation of three buildings to provide additional laboratories for the Chemistry Department.
- Relocation and conversion of a building for use by the Calibration Group.
- Renovation of a building to provide additional laboratories for the Biology Department.
- Repair of the Graphite Research Reactor cooling towers.
- Demolition of a low-pressure boiler plant.
- Exterior waterproofing of the heavy Machine Shop, Library, Cyclotron, Utilities-Maintenance, Hydrogen Liquefier, Waste Concentration, and Waste Disposal Buildings, and the exhaust ducts on the Graphite Research Reactor.
- Reroofing of four scientific buildings and three service buildings.

### SUPPLY AND MATERIEL

The continued expansion in the scope and diversity of the Laboratory's research programs

during fiscal 1960 was again reflected in an increase in the activity of the Supply and Materiel Division required to support them. The materiel handling activity, including receiving, storing and distributing supplies and equipment, increased  $\approx 12\%$ , based on the dollar volume of materiel handled during the year.

The materiel control activity, including the operation of stockrooms, inventory control, and property management, increased  $\approx 13\%$ , based on the dollar value of the inventory during this period. At the same time the inventory turnover was sufficient to reduce the investment in active inventory to 3.2 months of the total annual value. This compares with a value of 3.35 months for the previous year. For detailed inventory changes see Table 4, Introduction.

During the year the property management activity included the completion of an inventory of Laboratory capital equipment in which the punched card records showing the number, description, account charge, and location of some 20,000 items were brought up to date. Extensive savings to the Laboratory were made through the acquisition for \$85,775 of excess Government material and equipment originally valued at \$1,678,061.

To further the aim of bringing supply facilities closer to users, two additional outlying stockrooms were established during the fiscal year.

### PURCHASING

The growth of the Laboratory's research programs in fiscal 1960 resulted in a substantial expansion of the activities of the Purchasing Division, as indicated by the  $\approx 12\%$  increase in the total dollar volume of material and services procured. A comparison of the procurement activities for fiscal 1959 and 1960 in terms of transactions and dollar volume is shown in Table 8.

It will be noted that the number of contract transactions more than tripled during the year, which made advisable the establishment within the Purchasing Division of a Contract Section in addition to the Stores Section and the Direct Buying Section.

Besides the expanded use of petty cash for purchases, further economies were effected by the use of blanket orders for the scheduled delivery of certain classes of stock items. The procedure for handling such orders was improved by combining the

Table 8

Procurement method	Fiscal 1960		Fiscal 1959	
	No. of transactions	Volume, \$	No. of transactions	Volume, \$
Purchase orders	19,398	9,462,445	18,140	8,493,000
Contracts	166	1,626,931	55	1,434,000
Petty cash	6,356	154,215	5,285	130,000
Total	25,920	11,243,591	23,480	10,057,000

vendor's invoice and packing list with a set of receiving forms, to be prepared by the vendor for each delivery. This eliminates the preparation by the Laboratory of any further orders, releases, or

receiving forms after the placement of the initial blanket order.

During fiscal 1960, twelve sales of surplus Laboratory property resulted in a return of \$37,227.76.





## Appendix A

### PUBLICATIONS, JULY 1, 1959 – JUNE 30, 1960

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, 1959. BNL 560 (AS-13).  
Progress Reports, Nuclear Engineering Department:  
January 1 – April 30, 1959. BNL 571 (S-52).  
May 1 – August 31, 1959. BNL 583 (S-53).  
September 1 – December 31, 1959. BNL 595 (S-54).  
Conference Reports:  
Brookhaven Symposia in Biology No. 12. *Structure and Function of Genetic Elements*. BNL 558 (C-29).  
Weekly Bulletin 12, No. 51-2; 13, No. 1-50.  
Weekly Selected Reading List 12, No. 17-52; 13, No. 1-15.

#### STAFF PUBLICATIONS AND ABSTRACTS

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- BLEWETT, J.P. *Application of the Linear Accelerator for Production of Intense Proton Beams at 10 Bev. II. Neutrino beams*. Informal Report BNL 4660/JPB-14.
- BLEWETT, J.P. The Brookhaven Alternating Gradient Synchrotron. 1960 IRE Intern. Convention Record, Part 9, pp. 3-10.
- BLEWETT, J.P. *The Focal Properties of Certain Quadrupole Lenses*. Informal Report BNL 4654/JPB-13.
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- BLEWETT, M.H. *Some Saturation Characteristics of Alternating Gradient Magnet Models*. Informal Report BNL 4640/MHB-6.
- BROWN, H.N. *Injection Optical System*. Informal Report BNL 4646/HNB-1.
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- COTTINGHAM, J.G. *Ripple Filtering for Main AGS Magnet Generator*. Informal Report BNL 4650/JGC-14.
- COURANT, E.D. *Beat Factors and Stopband Widths*. Informal Report BNL 4641/EDC-24.
- COURANT, E.D. *Quadrupole and Sextupole Requirements*. Informal Report BNL 4644/EDC-26.
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- GIORDANO, S. The linear accelerator injector for the AGS. 1960 IRE Intern. Convention Record, Part 9, pp. 11-18.
- GREEN, G.K. *Cosmotron Magnet Power Supply*. Informal Report BNL 4632/GKG-1.
- HART, J.W. *Alignment of AGS Linear Accelerator Tank*. Informal Report BNL 4655/JWH-1.
- KEVEY, A. *Drift Tube Alignment for the AGS Linac*. Informal Report BNL 4689/AK-1.
- KIESLING, J.D. *Linac Tuning System*. Informal Report BNL 4645/JDK-2.
- KIESLING, J.D. *Modes in Long Cavities*. Informal Report BNL 4652/JDK-6.
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\*Also included are those listings from the last Annual Report [BNL 560 (AS-13)] for which complete reference information was not then available.

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- PALMER, J. P. *The Dynamic K-Meter*. Informal Report BNL 4657/JPP-9.
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- POLK, I. J. *Linear Accelerator Drift Tube Support*. Informal Report BNL 4634/IJP-4.
- RAKA, E. C. *Beam Observation Electrodes for the AGS*. Informal Report BNL 4643/ECR-3.
- READING, O. S. *Interim Data on the Stability of AGS Foundations*. Informal Report BNL 4661/OSR-4.
- RHEAUME, R. H. *A Parallel-Transistor Cascaded Amplifier for Controlling Very Large Currents*. Informal Report BNL 4651/RHR-5.
- RUTAN, E. J. *Saturating Inductor for Controlling the Magnetic Field of the Alternating Gradient Synchrotron*. Informal Report BNL 4659/EJR-8.
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- SMITH, L. *Orbit Considerations in the Linear Accelerator*. Informal Report BNL 4635/LS-3.
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Dec.	1959	BNL 589 (T-167)
Jan.	1960	BNL 591 (T-168)
Feb.	1960	BNL 597 (T-171)
Mar.	1960	BNL 601 (T-174)
Apr.	1960	BNL 604 (T-176)
May	1960	BNL 611 (T-181)
June	1960	BNL 620 (T-188)

POWELL, R.W. - See also ROBERTSON, J.S. (Medical).



## 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\**  
 Charles E. Falk, *Assistant to the Director*  
 Samuel M. Tucker, *Assistant to the Director*

George K. Green, *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  
 Irving J. Polk, *Head*, Mechanical Engineering Department  
 Lee E. Farr, *Chairman*, Medical Department  
 Clarke Williams, *Chairman*, Nuclear Engineering Department  
 Maurice Goldhaber, *Chairman*, Physics Department  
 Robert W. Powell, *Head*, Reactor Division

J. Georges Peter, *Director*, Architectural Planning  
 H. Russell Cort, *Budget Officer*  
 Lewis R. Burchill, *Controller*  
 Dennis Puleston, *Information Officer*  
 Charles F. Dunbar, *Legal Counsel*  
 Joseph S. Washburne, *Personnel Manager*  
 Frederick H. Williams, *Security Officer*

#### Accelerator Development Department

George K. Green, *Chairman*  
 Roger R. Adams  
 Richard A. Beth  
 John W. Bittner  
 John P. Blewett  
 M. Hildred Blewett  
 Hugh N. Brown  
 Theodore N. Constant  
 James G. Cottingham  
 Gordon T. Danby  
 Donald A. Davis  
 Robert J. Gerrity\*  
 Salvatore T. Giordano  
 John W. Hart\*  
 (assigned from Lawrence Radiation  
 Laboratory)  
 Ralph R. Kassner  
 John D. Kiesling\*  
 John Lancaster  
 (assigned from Director's Office)

William T. Link  
 Robert A. Loper  
 Thomas F. Madigan  
 Lowell McLean  
 Walter W. Merkle  
 Robert H. Phillips  
 Martin Plotkin  
 Herman C. Praddaude  
 (IAEA Fellow)  
 Eugene C. Raka  
 Raymond H. Rheume  
 Everett J. Rutan  
 Edward E. Shelton  
 Julius Spiro  
 Arie Van Steenberg  
 John J. Walters

#### Applied Mathematics Division

Milton E. Rose, *Head*  
 Bruno F. Dejon\*  
 (postdoctoral appointment)  
 David L. Fox\*  
 (part time from New York Univ.)  
 Ray C. Makino\*

Martin Milgram  
 Joel D. Pincus  
 (postdoctoral appointment)  
 Stuart S. Rideout  
 Yoshio Shimamoto  
 Kenneth Smith\*  
 Gerson H. Sparer\*  
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#### Biology Department

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 David T. Armstrong  
 (postdoctoral appointment)  
 Leroy G. Augenstine  
 (on leave to AEC Division  
 of Biology and Medicine)  
 Chauncey R. Benedict  
 (postdoctoral appointment)  
 John Berech, Jr.  
 John A. Bergeron  
 James L. Brewbaker  
 Cornelis Broertjes  
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 in Agriculture, Netherlands)

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Samuel F. Conti  
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Phelps P. Crump\*  
Kevin R. Daly  
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R. Clinton Fuller  
(*on leave to Oxford Univ., England*)  
Eric C. Gaetjens  
(*postdoctoral appointment*)  
Anne D. Gounaris  
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C.H.W. Hirs  
James R. Innes  
James R. Klein  
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Marian E. Koshland  
Leo E. LaChance  
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Myron Levine  
Sanat K. Majumder\*  
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Miodrag M. Maric\*  
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Leslie F. Nims  
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Erwin A. Schwinghamer  
Seymour Shapiro  
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*Grenoble, France*)  
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Lewis Friedman  
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Jerome Hudis  
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Sydney O. Thompson  
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Alfred P. Wolf  
Max Wolfsberg  
John Yang\*  
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 George B. Chadwick  
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*(graduate student from  
 Carnegie Inst. of Technology)*  
 Lloyd G. Hyman  
*(assigned from Harvard Univ.)*  
 Joseph Keren  
*(graduate student from Columbia Univ.)*  
 Lawrence B. Leipuner  
 Isador J. Livant  
 Eugene C. Loh  
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 Massachusetts Inst. of Technology)*  
 Hugh J. Martin, Jr.  
*(on leave from Indiana Univ.)*  
 William H. Moore, Jr.  
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 Frank T. Shively  
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 Raul Brenner\*  
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 Inst. de Energía Atómica, Brazil)*  
 Arland L. Carsten  
 Robert L. Chase  
 Ching T. Chen-Tsai  
*(IAEA Fellow)*  
 Frederick P. Cowan  
 Carl H. Distenfeld  
 Joachim Fischer

\*Terminated before June 30, 1960.

Charles F. Foelix  
 Lee Gemmell  
 Thomas M. Gerusky\*  
 John S. Handloser  
 William J. Hartin  
 William A. Higinbotham  
 Roland P. Kenschaft\*  
 Charles B. Meinhold  
 Gabriel L. Miller  
 Thomas D. Murphy\*  
 James P. Palmer  
 Leigh F. Phillips  
 David W. Potter  
 John P. Potter\*  
*(graduate student from Univ. of Rochester)*  
 Ernest A. Rainey\*  
 Seymour Rankowitz  
 George E. Schwender  
 Irving A. Singer  
 Maynard E. Smith  
 Robert J. Spinrad  
*(on leave to  
 Massachusetts Inst. of Technology)*  
 Raymond W. Stong  
 Helge I. Strand\*  
*(on leave from Joint Establishment  
 for Nuclear Energy Research, Norway)*  
 Robert M. Sugarman  
 Vito A. Svelto\*  
*(assigned from Centro Informazioni  
 Studi Esperienze, Italy)*

#### **Mechanical Engineering Division**

Irving J. Polk, *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  
 Jack E. Jensen  
 Michael B. Karelitz  
 David A. Kassner  
 Calman Lasky\*  
 Robert J. McCracken  
 Kurt F. Minati  
 George Nugent  
 Oliver S. Reading  
 Albert P. Schlafke, Jr.  
 Charles Theisen  
 William G. Walker

#### **Medical Department**

Lee E. Farr, *Chairman*  
 Gonzalo E. Aponte  
*(assigned from Jefferson Medical College)*  
 John L. Bateman  
*(medical associate)*

Albert Bertinchamps\*  
*(medical associate)*  
 Victor P. Bond  
 Donald C. Borg  
 Wenceslao Calvo  
*(on leave from Facultad de Medicina,  
 Valencia, Spain)*  
 Stanton H. Cohn  
 Spencer L. Commerford  
*(assigned from National Cancer Inst.,  
 National Inst. of Health)*  
 Robert A. Conard, Jr.  
 George C. Cotzias  
 Robert L. Cranny\*  
*(medical associate)*  
 Eugene P. Cronkite  
 Lewis K. Dahl  
 Albert F. Debons  
*(medical associate)*  
 Roger C. De Meutter  
*(medical associate)*  
 Ruth M. Drew  
 Otho D. Easterday  
 Ludwig E. Feinendegen  
*(medical associate)*  
 Samuel Fine  
*(medical associate)*  
 Conrad Fong  
*(assigned from  
 New York Univ. School of Medicine)*  
 Olav P. Foss\*  
*(on leave from Norwegian Radiumhospital,  
 Oslo, Norway)*  
 William H. Gordon\*  
*(medical associate)*  
 Francine Gregoire  
*(assigned from  
 Scientific Commission of NATO)*  
 Mary B. Hagamen\*  
 Richard A. Hammerstrom  
*(medical associate)*  
 Lawrence V. Hanks  
 Edwin R. Hughes  
*(medical associate)*  
 Walter L. Hughes  
 Horton A. Johnson\*  
*(medical associate)*  
 Sven-Aage Killmann  
*(medical associate)*  
 Robert C. Krueger  
*(on leave from Univ. of Cincinnati)*  
 Paul G. LeFevre\*  
 Stuart W. Lippincott  
 Robert A. Love  
 Myles Maxfield\*  
 Nicholas Odartchenko  
*(medical associate)*  
 Paul S. Papavasiliou  
*(medical associate)*  
 Victor Perman\*  
*(medical associate)*  
 Edwin A. Popenoe

Peter G. Reizenstein  
(medical associate)  
James S. Robertson  
Joseph R. Rubini\*  
(medical associate)  
Irving L. Schwartz  
Claire J. Shellabarger\*  
Walton W. Shreeve  
Lawrence Silver  
(medical associate)  
Malcolm G. Smilay  
(medical associate)  
Sanford C. Spraragen  
(medical associate)  
Elmer E. Stickley  
Richard D. Stoner  
Geronimo Terres, Jr.  
Edgar A. Tonna  
(medical associate)  
Akira Tsuya\*  
(medical associate)  
Edward Usenik  
(on leave from Univ. of Wisconsin)  
Donald D. Van Slyke  
William Wolins  
Yasokazu Yamamoto  
(medical associate)

#### Nuclear Engineering Department

Clarke Williams, Chairman  
George Adler  
Leonard B. Adler  
Clemens Auerbach  
Theodore Auerbach\*  
Allan Auskern  
David S. Ballantine  
Conrad G. Baumann  
(assigned from S.B. Penick & Co.)  
Bahattin M. Baysal\*  
(on leave from Univ. of Ankara, Turkey)  
Fritz Bloch  
Joseph S. Bryner  
Charles Carlson  
Jack Chernick  
Joe G.Y. Chow  
Noel R. Corngold  
Anita J. Court  
Charles G. Dan\*  
(assigned from Babcock & Wilcox Co.)  
James R. Day, Jr.\*  
(assigned from Babcock & Wilcox Co.)  
Herbert J. Diener\*  
(assigned from Babcock & Wilcox Co.)  
Robert F. Doering  
Kenneth W. Downes  
Orrington E. Dwyer  
James J. Egan  
Ali M. Elatrash\*  
(on leave from Florida State Univ.)

Terminated before June 30, 1960.

Enis Erdik\*  
(on leave from Univ. of Ankara, Turkey)  
Allen Eshaya  
Harmon L. Finston  
Albert H. Fleitman  
Aaron J. Friedland\*  
(graduate student from Columbia Univ.)  
Yuzo Fukai  
(assigned from Nippon  
Atomic Industry Group Co., Japan)  
Althea Glines  
David H. Gurinsky  
Loranus P. Hatch  
Aulis A. Hellsten\*  
(assigned from  
State Power Co., Finland)  
Joseph Hendrie  
Raymond J. Heus  
Frank B. Hill  
Manny Hillman  
Jurg A. Hoigne\*  
(postdoctoral appointment)  
Henry C. Honeck  
Frederick L. Horn  
Robert J. Isler  
Toshio Iwaki  
(assigned from Mitsubishi  
Atomic Power Industries, Inc., Japan)  
William T. Johnsen\*  
(assigned from Babcock & Wilcox Co.)  
Richard Johnson  
Sheldon Kalish  
Otto F. Kammerer  
Kouichiro Kashiwabara\*  
(assigned from Mitsubishi  
Atomic Power Industries, Inc., Japan)  
Herbert M. Katz  
John J. Kelsch  
William F. Kenney  
Carl J. Klamut  
Shirley E. Knight\*  
(assigned from Babcock & Wilcox Co.)  
Juan U. Kopell  
(postdoctoral appointment)  
Herbert J.C. Kouts  
Otto Kuhl  
Gerald S. Lellouche  
Melvin M. Levine  
Donald R. MacKenzie  
Bernard Manowitz  
Michael W. Maresca  
Frank D. Maslan  
Owen E. McCoy\*  
(assigned from Babcock & Wilcox Co.)  
William E. McNulty\*  
Donald J. Metz  
Robert A. Meyer\*  
Paul A. Michael  
Francis T. Miles  
Edmund R. Modowski\*  
(assigned from Babcock & Wilcox Co.)  
Leonard Newman

Granville M. Olds\*  
(assigned from Babcock & Wilcox Co.)  
Hassan Parnianpour\*  
(on leave from  
Bagdad Pact Nuclear Centre, Iraq)  
James P. Phelps  
Carl Pierce  
James R. Powell, Jr.  
Glen A. Price  
Aldyr A. Quadrado  
(assigned from Brazilian Army, Brazil)  
Venkatesalu Rajagopal  
(graduate student from  
Rensselaer Polytechnic Inst.)  
Chad J. Raseman  
William H. Regan, Jr.  
James J. Reilly, Jr.  
Powell Richards  
Herbert W. Rief\*  
(postdoctoral appointment)  
Francis X. Rizzo  
William A. Robba  
Wolfgang Rothenstein\*  
(postdoctoral appointment)  
Jerome Sadofsky  
Francis J. Salzano  
César A. Sastre  
Clifford H. Scarlett  
Donald G. Schweitzer  
Thomas V. Sheehan  
Rudolph Sher  
Nagao Shibata\*  
(on leave from Japan  
Atomic Energy Research Inst., Japan)  
Yasuo Shinohara  
(on leave from Tokyo Rayon Co., Japan)  
Robert M. Singer  
Louis M. Slater  
Maxwell M. Small  
(assigned from  
Director's Office for HFBR project)  
John L. Speirs  
Louis G. Stang, Jr.  
Eugene Starr  
Meyer Steinberg  
Gerald Strickland  
Herbert Susskind  
Hiroshi Takahashi  
(postdoctoral appointment)  
Stelvio Tassan  
(graduate student from New York Univ.)  
Walter D. Tucker  
John D. Van Norman  
(postdoctoral appointment)  
Jean I. Wagner  
Charles H. Waide  
Robert J. Walther\*  
(assigned from Babcock & Wilcox Co.)  
John R. Weeks  
Eugene V. Weinstock  
Jerome Weiss  
Henry H. Windsor

Edward Wirsing, Jr.  
 Richard H. Wiswall, Jr.  
 Emanuel Yellin  
*(postdoctoral appointment)*  
 Shigekazu Yoshijima  
*(assigned from Nippon Atomic Industry Group Co., Japan)*  
 Samuel A. Zwickler\*

#### Physics Department

Maurice Goldhaber, *Chairman*  
 David E. Alburger  
 Louis C.R. Alfred  
*(postdoctoral appointment)*  
 Harvey A. Alperin  
*(assigned from U.S. Naval Ordnance Laboratory)*  
 Frank Anderson  
*(on leave from The Johns Hopkins Univ.)*  
 Aurelio Ascoli  
*(assigned from Centro Informazioni Studi Esperienze, Italy)*  
 Frederick Ayer II  
 Charles P. Baker  
 Winslow F. Baker  
 Herbert H. Bolotin  
*(postdoctoral appointment)*  
 Ernest M. Bolze  
 Morton K. Brussel\*  
 Robert E. Chrien  
 Eugene L. Church\*  
*(assigned from Frankford Arsenal)*  
 Victor W. Cohen  
 Derek C. Colley  
 Rodney L. Cool  
 Hans W. Courant\*  
*(on leave from Yale Univ.)*  
 Bernard B. Culwick  
 Arthur C. Damask  
*(assigned from Frankford Arsenal)*  
 Edward der Mateosian  
 George J. Dienes  
 Loyal Durand III  
*(on leave to Univ. of Colorado)*  
 Guy T. Emery  
*(postdoctoral appointment)*  
 William J. Fickinger  
*(graduate student from Yale Univ.)*  
 Nikolaus F. Fiebiger  
*(on leave from Univ. of Frankfurt)*  
 Ted B. Flanagan  
 Hugie L. Foote, Jr.\*  
*(on leave from Bell Telephone Laboratories)*  
 William B. Fowler  
 John D. Fox\*  
 William M. Frank\*  
*(postdoctoral appointment)*  
 B. Chalmers Frazer

André Gallmann\*  
*(postdoctoral appointment)*  
 Orn S. Gardarsson  
*(IAEA Fellow)*  
 John B. Gibson  
 G. Norris Glasoe  
 Allen N. Goland  
*(assigned from Watertown Arsenal)*  
 Gertrude S. Goldhaber  
 Samuel A. Goudsmit  
 Edward L. Hart  
 John W. Hess  
*(assigned from Picatinny Arsenal)*  
 John Hornbostel  
 Donald J. Hughes  
*(deceased April 12, 1960)*  
 Joseph Jach  
*(assigned from Picatinny Arsenal)*  
 Adeshwar Jain  
*(graduate student from Cornell Univ.)*  
 Edgar W. Jenkins  
*(postdoctoral appointment)*  
 H.K. Alan Kan  
*(assigned from Picatinny Arsenal)*  
 John V. Kane  
 Walter R. Kane  
*(postdoctoral appointment)*  
 Mortimer I. Kay\*  
*(on leave from Pennsylvania State Univ.)*  
 David T. Keating  
 Vincent P. Kenney\*  
*(on leave from Univ. of Kentucky)*  
 Edward H. Kerner\*  
*(on leave from Univ. of Buffalo)*  
 Ottmar C. Kistner  
*(postdoctoral appointment)*  
 Walter Kley\*  
*(on leave from Albert Ludwigs Universität, Freiburg, West Germany)*  
 Joshua K. Kopp  
 Henry L. Kraybill  
*(on leave from Yale Univ.)*  
 Thaddeus F. Kycia  
*(postdoctoral appointment)*  
 Leon F. Landovitz  
*(postdoctoral appointment)*  
 Robert E. Lanou, Jr.  
*(assigned from Brown Univ.)*  
 Ronald E. Larsen  
*(assigned from Frankford Arsenal)*  
 Paul J. Leurgans  
*(assigned from The Physical Review)*  
 Paul W. Levy  
 Seymour J. Lindenbaum  
 Robert I. Louttit  
 Dierk Luers  
*(on leave from Max Planck Inst., Munich, West Germany)*  
 James E. Mapes  
*(assigned from Picatinny Arsenal)*  
 Robert B. Marr  
*(postdoctoral appointment)*

Harvey Marshak  
 Leona W. Marshall  
*(on leave from Univ. of Chicago)*  
 Joseph P. Martin  
*(postdoctoral appointment)*  
 Werner A.W. Mehlhop  
*(postdoctoral appointment)*  
 Adrian C. Melissinos  
*(assigned from Univ. of Rochester)*  
 Toshio Mitsui\*  
*(on leave from Pennsylvania State Univ.)*  
 Hans B. Möller\*  
*(on leave from Danish Atomic Energy Commission, Denmark)*  
 James A. Moore  
*(assigned from Columbia Univ.)*  
 Thomas W. Morris  
 Bernard Mozer  
*(postdoctoral appointment)*  
 Francis Muller  
*(on leave from Lawrence Radiation Laboratory)*  
 Robert Nathans  
 James A. Niederer  
 Shimon Ofer\*  
*(postdoctoral appointment)*  
 Satoshi Ozaki  
*(postdoctoral appointment)*  
 Harry Palevsky  
 Simon Pasternak  
*(assistant editor of The Physical Review)*  
 François Penet  
*(assigned from Centre d'Études Nucléaires, Saclay France)*  
 Oreste Piccioni  
 Stanley J. Pickart  
*(assigned from U.S. Naval Ordnance Laboratory)*  
 Eric Pickup  
*(assigned from National Research Council, Canada)*  
 Ralph A. Pixley  
*(postdoctoral appointment)*  
 Albert G. Prodel  
 David C. Rahm  
*(on leave to Centre d'Études Nucléaires, Saclay, France)*  
 Ralph R. Rau  
 Charles A. Reynolds\*  
*(on leave from Univ. of Connecticut)*  
 Donald K. Robinson  
*(postdoctoral appointment)*  
 Ronald M. Rockmore  
*(postdoctoral appointment)*  
 David B. Rosenblatt  
*(assigned from Frankford Arsenal)*  
 John J. Russell, Jr.  
*(postdoctoral appointment)*  
 Brice M. Rustad  
*(assigned from Columbia Univ.)*  
 George J. Safford  
*(assigned from Columbia Univ.)*

\*Terminated before June 30, 1960.

Vance L. Sailor  
 Edward O. Salant  
 Nicholas P. Samios  
 James R. Sanford  
*(graduate student from Yale Univ.)*  
 Jack Schwartz  
*(on leave from Radio Corp. of America)*  
 Robert B. Schwartz  
 Arthur Z. Schwarzschild  
 Ferdinard J. Shore, Jr.  
 Ralph P. Shutt  
 Joseph E. Smith  
 Hartland S. Synder  
 Andrew W. Sunyar  
 John B. Swan  
*(on leave from*  
*Univ. of Western Australia, Nedlands)*  
 Louis J. Teutonico\*  
*(assigned from Brown Univ.)*

Allan Thorndike  
 Ryuzo Ueda\*  
*(on leave from Waseda Univ., Japan)*  
 George H. Vineyard  
 Joseph B. Vise  
*(graduate student from Columbia Univ.)*  
 Howard W. Wagenblast  
*(assigned from U.S. Steel Corp.)*  
 Yoshinisa Wakuta  
*(on leave from Kyushu Univ., Japan)*  
 Joseph Weneser  
 Gian Carlo Wick  
 William J. Willis  
 Sukeyasu S. Yamamoto  
 David A. Young\*  
*(postdoctoral appointment)*  
 Robert L. Zimmerman\*  
 Gus T. Zorn  
 Martin S. Zucker

#### Reactor Division

Robert W. Powell, *Head*  
 Sidney C. Abrahams  
*(assigned from Bell Telephone Co.)*  
 John E. Binns  
 John J. Floyd  
 Gerald C. Kinne  
 Issai Lefkowitz\*  
*(assigned from Glenco Corp.)*  
 Donald E. Neil\*  
*(assigned from*  
*Rensselaer Polytechnic Inst.)*  
 Willem F. Oosterheert  
*(on leave from Inst. for Atomic Sciences*  
*in Agriculture, Netherlands)*  
 Charles L. Osborne  
 Jack E. Phillips  
 dePuyster G. Pitcher  
 Harry H. Steinhauser, Jr.  
*(assigned from*  
*Rensselaer Polytechnic Inst.)*

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\*Terminated before June 30, 1960.