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BNL 51 (S-5)

BROOKHAVEN NATIONAL LABORATORY

Quarterly Progress Report

January 1 - March 31, 1950

Associated Universities Incorporated
under contract with the
United States Atomic Energy Commission

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Printed at Upton, New York, for distribution
to individuals and organizations associated
with the national atomic energy program.

April 1, 1950 500 copies

INTRODUCTION

This is the first of a series of Quarterly Reports. These reports will deal primarily with the progress made in our scientific program during a three months period. Those interested in matters pertaining to organization, administration, complete scientific program, personnel and other matters not directly involved in current scientific progress are referred to our Annual Progress Report which is issued in January.

We have attempted to describe new information that appears significant, or of interest, to other scientists within the Atomic Energy Commission Laboratories. No effort has been made, however, to detail progress in each and every research project. Little or no reference will therefore be found to the projects in which progress during the current period is considered too inconclusive.

Since our organizational structure is departmental, the work described herein is arranged in the following sequence:

Accelerator Project
Biology Department
Chemistry Department
Instrumentation and Health Physics Department
Medical Department
Physics Department
Reactor Science and Engineering Department

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ACCELERATOR PROJECT

THE COSMOTRON

During the period covered by this report progress of the Cosmotron has been approximately according to schedule. The machine is out of the paper stage and all the major components are beginning to exist physically with the exception of the vacuum system. The broad physical design of the machine is complete and the important parameters have all been fixed. There follows a report of the progress made on the various components. For those not familiar with the project, the reader is referred to a review article by Livingston, et al, Review of Scientific Instruments, January 1950.

Building and Power Supply

The structure of the building is now completed and the contractor has installed half of the building services and auxiliary devices. The two 10-ton, 60 ft. span cranes were installed and have been used a great deal during this period. The building was closed to the weather and heated temporarily so that numerous projects could be carried on during the cold weather. The transformer sub-station was received and installed on its foundation, but is not yet wired. All of the motor generator set and many of the other components of the Westinghouse power supply system have been received; the bearings and rotating parts of the machine are in the building and are now in the process of erection.

Magnet

The major advance during this period has been the erection of the magnet steel. The magnet bed plates were installed and leveled and the 288 magnet blocks have been loaded on these bed plates and adjusted. Over half are now welded in place. The foundation for these blocks so far seems quite stable and, by careful shimming, the median planes of all blocks lie in a plane within better than .010". Other tolerances on the structure have been held to corresponding limits.

An intensive magnet test program in which each of the 6-ton magnet blocks was inserted in a fixture and run through a complete set of magnetic measurements was completed. Some 8,000 oscillographs were analyzed to obtain over 150,000 data points. A statistical reduction of these data was then used to determine the order in which the blocks were set down in the ring. By this process it is anticipated that we can keep the first, second and third order magnetic variations to less than 1 part in 1,000.

Coil

The final details of design of the coil and its insulation have been worked out by the construction of test samples and engineering tests on them. Fabrication of the first quadrant coil has been started and over half of the bar insulation for the whole job has been applied.

Radio Frequency System

The master oscillator of the radio frequency system has undergone further refinement and intensive testing so that we are now able to control this oscillator over a range from 250 kc to 4.5 mc with a precision and repeatability somewhat better than 0.1 percent. Intermediate stages have been breadboarded and the final high power stages are under construction. The power supply for the final radio frequency stage (300 kw d-c) is in place but has not been wired. Ten of the ferrite laminations for the radio frequency core have been received from Philips. Tests indicate that the quality of these, as well as General Ceramics Company material, are of somewhat better characteristics than our original specifications. The various components of the analogue computing system which tracks the radio frequency with the varying magnetic field have been tested separately and each satisfied the precision requirements.

Wiring and Controls

Engineering design of the control system for the machine as a whole has progressed to the point where wiring runs have been laid out and the very detailed design is started. The complexity of the task can be seen from the fact that between 2,000 and 3,000 wires will come from all parts of the machine into the control room.

Vacuum System

The vacuum system is the one remaining major component for which a final design is lacking. Intensive testing of sample structures and various materials was carried out and a considerable amount of important data has been collected. Two general designs are under consideration. An all metal chamber which incorporates 10 mil Inconel foil skin seems most satisfactory from the standpoint of obtaining a low pressure. This design suffers from the fact that the metal skin introduces additional eddy currents which complicate the problem of obtaining the proper magnetic field in the magnet gap. To avoid this difficulty test chambers have been constructed which incorporate plastic skins. Although these plastic skins are free from eddy currents, they have not been completely proved with respect to their vacuum properties. The testing has progressed to the point where a number of designs seem possible. The question of which is the best has yet to be determined. A prototype pump unit has been constructed; most of the parts for 15 complete systems are on hand, and construction of the 15 pump units is under way.

Injection System

The injection Van de Graaff has been finished by our contractor, the High Voltage Engineering Company, and has run satisfactorily at 4.8 Mev without a beam. Completion of this contract is now delayed by difficulties in the accelerating tube which presently limit the voltage, but steps are being taken to correct this difficulty which seems to arise in a secondary emission from the electrodes of the accelerating tube. The injection system which is to launch the beam into the proper orbits has been designed and numerous parts have been fabricated. This entire injection system, including the Van de Graaff generator and the ion optical system, must preserve a focus of the ion beam within angular limits of the order of .001 radians.

Miscellaneous

The theoretical group has made elaborate and fairly precise calculations on the expected influence on the beam of factors such as gas pressure, magnetic field inhomogeneities, etc. This study has provided important information with regard to tolerances on the magnetic field, radio frequency, and injection system.

Twenty-three internal project reports have been prepared and distributed to members of the project and to other interested laboratories. Since these reports are often tentative, their distribution has been restricted.

During the past three months the following papers have been published: "Design Study for a Three Bev Proton Accelerator", M.S. Livingston, J.P. Blewett, G. K. Green, and L. J. Haworth, R.S.I., January 1950, and "The Neutron Density in the Free Atmosphere up to 100,000 Feet" Luke C. L. Yuan, Phys. Rev., March 1, 1950.

THE GENERAL ELECTRIC VAN DE GRAAFF

This generator is running with a proton beam of 5μa at voltages from .75 to 3.0 Mev. The energy scale has been calibrated with respect to the 994 Kev resonance in aluminum and linearity has been checked with the 1901 Kev resonance. The machine regulates in this region by feeding back through a corona system with a fluctuation of less than 2 Kev. Voltage tests indicate that 3 Mev may be the upper limit to be obtained with this machine.

THE 60" CYCLOTRON

The stage has been reached where attempts to obtain a beam may begin in a few weeks. The magnet has been run in and a field in excess of 18,000 gauss obtained. The Dees have been inserted into the vacuum chamber and a good vacuum has been obtained. The oscillator is being tested.

BIOLOGY DEPARTMENT

The work of the Biology Department has been greatly facilitated by the completion of the first phase of the new laboratories. Specialized equipment, including an ultracentrifuge and an electron microscope, have been installed and are operating. An electrophoresis machine will be available soon. The installation of the new x-ray equipment has been postponed until late in the year. The new machines will make possible quantitative studies of the interaction of high energy radiations with large molecules, such as proteins, enzymes and nucleic acids obtained either from *in vivo* or *in vitro* experiments.

Two main fields of research are being pursued in the Biology Department. The first is concerned with experiments designed to increase our fundamental knowledge of the processes and mechanisms by which radiation affects living systems. Such studies also have the additional benefit of giving increased insight into the basic facts of living systems themselves. The choice of suitable living systems for the study of radiobiology and the observations to be made are matters that require careful consideration. It is only to be expected that fullest understanding will be achieved by examinations of the biological effects of irradiation in many varieties of plant and animal life from the simplest unicellular organisms to the most complex forms. At present, members of the Department are investigating physiological and biochemical consequences of radiation in rats and selected plants; cytological and cytochemical disturbances produced by radiation in the pollen mother cells of *Trillium* and *Tradescantia* and the genetic effects of radiation on corn and *E. coli*.

The second field of research makes use of radioactive isotopes as tracers in various metabolic and physiological problems. These range in extent from the use of carbon-14 in the study of biosynthetic and photosynthetic processes in plants to the systematic study of the organisms that accumulate and concentrate various chemical elements. The last problem is of importance in any consideration of the disposal of radioactive wastes, for the ultimate evaluation of the hazard incurred in the release of a particular radioactive element or compound depends upon precise knowledge of whether some living system has the ability to concentrate the material to dangerous levels.

BIOLOGICAL EFFECTS OF RADIATION

Animals:

The repose of mammals to whole body irradiation resembles in many ways the effects resulting from other forms of stress, such as exercise, low and high temperatures, anoxia, infection, etc. The relationship is most evident when the pituitary-adrenal system is studied.

"The Adrenal Gland", F. A. Hartman and K. A. Brownell. Published by Lea and Febiger, Philadelphia, 1949.

In experiments designed to test the direct participation of the adrenal gland in radiation response, it was found that shielding of the adrenal gland of rats exposed to whole body x-irradiation greatly increased the survival rate and that substitution therapy would decrease the mortality rates of irradiated adrenalectromized rats.

A. Edelmann, Federation Proc. 9, 36, 1950.

Other studies of the biochemical changes mediated through the pituitary-adrenal system in part, show that liver glycogen of fasted irradiated rats rises to a maximum at two days after exposure only to fall rapidly

on the third day and then remains near the levels the control animals exhibit for the remainder of the seven day observation period. The urinary nitrogen excretion of irradiated rats is depressed slightly below normal levels for several days after exposure when the exposure is near lethal levels.
(A. Edelmann and L. F. Nims)

Plants:

Studies have shown that there is at least a 50-fold change in sensitivity at various stages of meiotic nuclear cycles in Trillium erectum to x-rays and attempts are now being made to correlate these changes in sensitivity with the concentrations of nucleic acids in the cells because of the known sensitivity of the nucleic acids to high energy radiation. The experiments to date indicate that synthesis of desoxyribose nucleic acid ceases during the two meiotic divisions. This may imply a correlation between synthesis of nucleic acid and sensitivity. When the rate of formation of nucleic acid is decreasing or has stopped, sensitivity to radiation is high.

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A. H. Sparrow, M. J. Moses and R. Steele. To be presented at American Association for Cancer Research, April 1950, Atlantic City, N. J.

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In the course of these investigations the Feulgen reaction is being critically studied and it has been found that in vitro the Feulgen reaction measures the loss of purines following acid hydrolysis of desoxyribose nucleic acid. In the cytological section, both the liberated purines and the degradation of the polynucleotides are measured.
(A. H. Sparrow and M. J. Moses).

In experiments on the effects of radiation on the growth of Katahdin and Green Mountain potatoes, a single dose of 300 r of x-rays does not decrease yield. At least 1200 r is necessary to produce a significant decrease in yield. Under conditions of continuous γ-irradiation, no effect on top growth of potatoes has been found at dose rates up to 80 r per day and total doses up to 8500 r. Tradescantia suffers some inhibition of top growth after receiving 400 r of γ-irradiation. No effect was seen at lower dosage. (A. H. Sparrow and E. Christensen).

At various times there have been reports that slight amounts of radiation will stimulate growth of plants. An experiment has been completed which was designed to test whether exposure of seed corn to x-rays and cobalt γ-rays would influence subsequent growth. Seeds were exposed to total doses of 250 to 2000 r, planted and the height of the plants determined at weekly intervals. The plantings were made in four different 4 x 4 Latin squares and both standard early field corn and two short hybrids were tested. No effect on growth was observed with the dosages employed.

In testing the viability of seeds after exposure to radiation, it is time consuming to wait for germination trials. The dye 2-3-5-triphenyltetrazolium chloride is known to stain viable embryonic tissues. Tests and refinement of techniques previously employed demonstrate the dye will not only provide a suitable and much more rapid indicator of seed viability after irradiation but that it will also allow a quantitative estimate of the amounts of succinic dehydrogenase in plant tissues. These methods will greatly expedite studies of radiation effects on seeds.

Preliminary results are now at hand of the genetic effects of low level γ-radiation on corn. The number of mutations observed appears to be higher than expected. Much more experimental work is necessary to settle the issue but it does appear that continuous radiation of growing plants may produce genetic effects at a quite different level of dosage than required for cells or gametes in a resting stage. This is understandable in light of

Sparrow's work which shows more than a 50-fold variation in the various stages of meiotic division. Continuous radiation is certain to catch these cells in growing corn at their most sensitive stage.

(W. R. Singleton, R. Van Reen and M. Koester).

The growth characteristics of E. coli in media made radioactive with P-32 are being intensively studied. It has been found that continuous radiation interferes with growth by increasing the duration of the lag phase. However, at lower radiation dosages, a great increase in mutation rate (Streptomycin resistance) can occur with apparently normal growth.

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B. A. Rubin and P. Steinglass, Proc. Soc. Am. Bacteriologists, May 1950.

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Some of the mutations of E. coli produced by irradiation require Streptomycin for growth. In spite of this requirement there is no evidence that the organisms metabolise Streptomycin. Detailed examination is being made of this apparently inexplicable finding.

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B. A. Rubin and P. Steinglas - to be published in Microbial Genetics Bulletin, April 1950.

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To make an analysis of these results, it is necessary to know the dosage received by irradiation in small volumes by the contained radioactive isotopes. A theoretical study of the problem has been completed in collaboration with P. I. Richards of the Physics Department which provides a quantitative calculation of the energy absorbed in small volume, when the range of the emitted β-particles resulting from radioactive decay is large compared to the volume. The results of these calculations will appear in Nucleonics in the form of graphs showing the relations between the various parameters. These graphs will greatly aid in obtaining more precise knowledge of the dosage the *E. coli* receive.

(B. A. Rubin).

TRACER STUDIES OF BIOCHEMICAL PROBLEMS

Animals:

Extensive work by many investigators has been done on the *in vitro* examination of the enzyme systems by which mammalian tissues acquire their metabolic energy from hydrolysis and oxidation of carbohydrates. With radioactive phosphorus it is possible to check some of the conclusions arrived at from the *in vitro* work. Work done here has shown that intact liver follows the postulated scheme in as far as it can be checked by radioactive phosphorus. To gain further information a study is now being made of alloxan produced diabetes in the rat. It is found that inorganic phosphate content of the livers of alloxan rats is some 50% higher than in normal controls. The level of glucose-1-phosphate is decreased and adenosine polyphosphate is increased. There is also a shift in ratio of ATP and ADP. In the normal rat the ratio is 70% ATP and 30% ADP while, in contrast, in the diabetic animals the ratio is 10% ATP and 90% ADP.

(J. Sacks).

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J. Sacks and B. Damast, Federation Proc. 9, 111, 1950.

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Extensive studies are being made of iron metabolism in mammals and birds and the use of tagged red cells to determine various physiological constants. In collaboration with a group of investigators at the College of Physicians and Surgeons, Columbia University, a critical comparison has been completed of three independent methods of estimating the blood volume in the dog. It has been found that volumes determined by the carbon monoxide method are 2% above the T-1824 dye method while the radioactive iron tagged red cell method is some 11% below that of the dye method.

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J. L. Nickerson, L. M. Sharpe, W. S. Root, T. C. Fleming and M. I. Gregerson, Federation Proc. 9, 94, 1950.

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In the rat, the use of the tagged red cell method for blood volumes gives values of only 4.83 per 100 gr. of body weight. Though the blood volume obtained here for the rat agrees quite well with that found by another group of investigators it is markedly less than the values found by plasma labeling.

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L. M. Sharpe, G. G. Culbreth, J. R. Klein, Federation Proc. 9, 226, 1950.

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The reasons for the lack of agreement among the various methods of estimating blood volume is not clear but it is quite evident that in small animals particularly, the tagged red cell method should be looked upon with suspicion. Tagged red cells have also been used to measure the residual blood volumes of exsanguinated and perfused tissues of the rat. It has been found exsanguination removes 34% of the blood in the whole carcass while perfusion lowers it to 5%. The ease with which blood is washed out of the various tissues differs greatly. Perfusion reduces the residual blood in the kidney, brain and heart by 30-fold or more but in the spleen only 25% of residual blood left after exsanguination is removable by perfusion.
(L. M. Sharpe, G. G. Culbreth and J. R. Klein).

Probably one of the most useful isotopes in the study of intermediary metabolism and disease states in man is carbon-14. Its long physical half-life presents a potential hazard that has severely limited its application.

Much animal work is necessary for the determination of the biological half-life of C-14 tagged compounds before radioactive carbon can be used with complete safety in man.

A study is being made of the fate of tagged sucrose in mice and it is found that doses of tagged sugar in the mouse equivalent to a dose of 1.0 curie in a man is without discernable radiological effect. Analysis of a number of tissues including bone of a mouse five weeks after the ingestion of 1/2 mc. of C-14 sucrose gives concentrations in the tissues ranging from 0.21 $m\mu c/mg$ C to 1.85 $m\mu c/mg$ C. The only tissues above 1.30 in activity are the abdominal fat, the brain and the blood. Forty to 50% of the radioactive carbon ingested in the form of sucrose is excreted in the first three hours. The factors that effect this elimination are also being studied. While the tagged CO_2 is somewhat variable in amount, the ratio of tagged CO_2 to total CO_2 is quite constant at comparable times after feeding and in a 24 gram mouse that has been fed 200 mg. of sucrose and 56 grams of amino acids, 50% of the expired CO_2 is derived from the sucrose 1-1/2 hours after feeding. If the same mouse is studied 4 to 7 weeks after the initial ingestion of tagged sucrose, it is found from the decrease in specific activity of the exhaled CO_2 after feeding normal sucrose and amino acids following a ten hour starvation period that 75% of the CO_2 is from "exogenous sources". The amino acids and sucrose are fed in the proportions found by Maddy and Elvehjem to be an adequate diet for the mouse.

(R. Steele).

Plants:

A general study of biosynthesis and photosynthesis in selected plants is being made. Such studies not only provide useful tagged compounds for other scientific work but also serve to increase our fundamental knowledge of the biochemistry of anabolic reactions in the plant. Some 15 other scientific studies are making use of sugars prepared here (sucrose, starch, fructose and glucose).

At one time it was generally believed that CO_2 was fixed by the plant in the form of formaldehyde which was then further condensed to form carbohydrates. This hypothesis is being tested by feeding tagged formaldehyde to preilluminated algae. If CO_2 has to pass through HCHO on its way to sugar or if the algae produce HCHO the specific activity of the fed $HC^{14}HO$ would have been reduced. No dilution was found, despite the fact the algae (*Scenedesmus*) did become radioactive. When sunflower leaves are exposed to tagged formaldehyde, some tagged CO_2 is produced. However the small amounts of formaldehyde utilized cannot account for the sugar produced. It seems certain in these preliminary experiments that though plants metabolize formaldehyde, the photosynthetic production of sugar does not proceed by a formaldehyde step.

To make the compounds obtained from plant sources most useful for scientific research, they must be extracted in pure state and the extent of labeling in each position determined with precision. There has been some question as to the mechanism of degradation when *Lactobacillus casei* is used to ferment glucose to lactic acid. A study of this fermentation shows that a partial randomization takes place between carbons 1 and 3 of glucose.

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M. Gibbs, R. Dumrose, F. A. Bennett and M. R. Bubeck. To be published in the June 1950 issue of J. Biol. Chem.

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The synthesis of labeled ascorbic acid has been completed. This involved the cooperation of both the biologists and the synthetic organic chemists and is more fully described in the report from the Chemistry Department.

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A. A. Bothner-By, M. Gibbs, R. C. Anderson. To be published in Science.

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(M. Gibbs).

Accumulator organisms:

Various living systems have the power of accumulating and concentrating elements to a high degree. The social Vespida, which comprise hornets, yellow jackets and the social wasps, absorb and retain virtually all of the manganese entering the digestive tract. In studies with radioactive tracers, it has been shown by chemical analyses that the manganese passes only into the cells of the mid intestine and remains there as long as the cells are alive. At the time of pupation, the larval intestine breaks down and the manganese is expelled from the body. Thus a newly emerged adult is almost as free of manganese as if he had never been exposed. The various adult castes in the Vespids differ in their manganese content. The present work shows that nutrition cannot be responsible for the sexual activity of the "worker" females after the adult stage is reached. Both males and fertile females, so long as they stay in the nest, are shown to eat food which differs from that of the larvae and workers. Thus, tracer studies and chemical analyses are of help in analyzing the structure of societies as well as that of organisms.

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V. T. Bowen. To be published in the J. of Exptl. Zoology.

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A hydroponics unit for the growth of plants in radioactive media has been completed and tested with arsenic-76 and phosphorus-32 on selected plants with the assistance of other members of the Department. This unit will be used to study the rare-earth accumulation in hickory seedlings. The hickory tree is the only organism that is known to accumulate rare-earths to any extent and the metabolism of the rare-earths in this plant is of great interest.

Other organisms under study include iron absorption in Drosophila, nickel and fluorine accumulation in a series of sponges and manganese-52 in fresh water clams. Such studies are not only of great interest from the waste disposal point of view but also from the general ecology of accumulator organisms. (V. T. Bowen).

Under the heading of General Chemistry of Various Elements are included (1) a report of work on magnetic properties of $MnCl_2$ and $MnCl_2$-$SrCl_2$ solid solutions, undertaken in order to reach a better understanding of the magnetic properties of uranium compounds as measured in this Laboratory, (2) some studies of optical and other properties of fluid solutions at liquid nitrogen temperature, and (3) some preliminary investigations of methods of recovery of protactinium 231 from various available residues.

No report of work in the field of Radiation Chemistry is included. The 2-Mev Van de Graaff accelerator has been received, on February 7, 1950, and most of the time of the radiation chemistry group since then has been taken up with the setting up and preliminary testing of this machine.

The study of Recoil Atoms and the Chemical Effects of Nuclear Transformations has been continued, and some recent results on the recoil of Be^7 atoms from surfaces following electron capture are reported.

The following Chemical Studies with Isotopes are described: (1) recent progress of work on the exchange reaction between thallous and thallic ions, (2) a preliminary report on the exchange between ferrous and ferric ions, (3) a mass-spectrometric study of the relative yields of atomic and molecular ions of hydrogen, deuterium, and tritium after electron impact, and (4) a rather complete investigation of isotope effects in the decomposition of ammonium nitrate into N_2O and H_2O, as well as some work on the vibration frequencies of isotopic N_2O molecules.

In the field of organic chemistry the successful synthesis of uniformly C^{14} labelled L-ascorbic acid (vitamin C) is reported.

Of the various nuclear problems under investigation the one which has reached a sufficient state of completion to be reported at this time is the study of the decay scheme of nickel 57.

THE MAGNETIC SUSCEPTIBILITY OF A SOLID SOLUTION OF MANGANOUS CHLORIDE AND STRONTIUM CHLORIDE

In connection with the measurement of the magnetic properties of compounds of the heavy elements, experiments are being carried out which attempt to give a clearer understanding of one of the important laws of magnetism, the Weiss-Curie law. This may be written

$$\chi = \frac{C}{T + \Delta}$$

where χ is the molar magnetic susceptibility, T is the absolute temperature, C is the Curie constant, and Δ the paramagnetic Curie temperature. Although the law is empirical and covers the magnetic behavior of a large class of salts over a wide temperature range, a completely satisfactory theoretical derivation has not yet been given. The various interpretations of the Weiss-Curie law are discussed by Van Vleck[1]. These center about the interpretation of the paramagnetic Curie temperature, which may be due to any of the following causes: (1) Heisenberg exchange between paramagnetic ions, (2) crystalline field Stark effect, and (3) dipole-dipole interaction between magnetic ions, this last effect being very small in general. By the proper choice of paramagnetic salts and the technique of diluting these with diamagnetic salts, it has become possible to sort out the various effects, and to study them independently[2].

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[1] J. H. Van Vleck, Theory of Electric and Magnetic Susceptibilities, Oxford Press

[2] Brookhaven Progress Report, July 1-Dec. 31, 1949, BNL 39, p. 48

An experiment was performed which attempted to bring about a change in the exchange interaction energy of paramagnetic ions in a crystal by a change in crystal structure. If such an effect existed, it would appear as a paramagnetic Curie temperature term in the Weiss-Curie law. It was not found however, but the experiment, which is of some interest, is reported here.

Manganous chloride, $MnCl_2$ has a magnetic susceptibility very nearly in accord with Curie's law for the free ion, $\chi = C/T$. It is actually given by the equation $\chi = 4.08/(T + 3.3^\circ)$.[3] This is unique among anhydrous chlorides of the iron group which generally have Δ terms larger by an order of magnitude than the one found for $MnCl_2$. The magnetic properties of manganous chloride might be associated with its unusual layer structure[4] built up of alternate single layers of manganese ions and double layers of chloride ions along the hexagonal axis. The double layer of chloride ions could act as an insulator for paramagnetic ion spin exchange, and so cause the Δ term nearly to vanish.

It was found possible to dissolve anhydrous $MnCl_2$ in anhydrous $SrCl_2$, which has the fluorite structure. These salts were prepared from C.P. materials and subsequently analyzed. A 30 mol % $MnCl_2$ - 70 mol % $SrCl_2$ mixture was fused at 800° C in an atmosphere of dry HCl. An X-ray powder photograph showed it to have the $SrCl_2$ structure with $a_0 = 6.95 Å$. For pure $SrCl_2$ $a_0 = 7.00 Å$.

The magnetic susceptibilities of pure $MnCl_2$ and of the mixture were then measured as a function of temperature. The results are shown in the graph. For pure $MnCl_2$ the susceptibility is given by the equation

$$\chi_{mol} = \frac{4.0}{T + 4^\circ}$$

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[3] Starr, Bitter and Kaufman, Phys. Rev. 58, 977 (1940)

[4] Wyckoff, The Structure of Crystals, Chemical Catalog Co., Inc. 1931, p. 233

and for $MnCl_2$ dissolved in $SrCl_2$ by the equation

$$\chi = \frac{4.28}{T}.$$

The moments calculated for the Mn^{++} ion are 5.68β and 5.92β respectively for the two cases. These results are in satisfactory agreement with the measurements of Starr, Bitter, and Kaufman[3] and with the calculated value for the moment, 5.92β for the ground state $^6S_{5/2}$.

The evidence suggests that spin-spin exchange cannot readily occur in chlorides, perhaps because of the large interatomic distances as compared to the case of oxides and fluorides. For those chlorides which do follow the Weiss-Curie law the Δ term may be entirely due to the crystalline field Stark effect. An experiment is planned to give further information on this point. (N. Elliott)

ULTRAVIOLET ABSORPTION SPECTRA OF SOME RARE EARTH SALTS IN SOLUTIONS FLUID AT THE TEMPERATURES OF LIQUID NITROGEN

Absorption spectra were taken of gadolinium bromide hexahydrate and also neodymium bromide hexahydrate dissolved at 77° K in one part propyl alcohol and four and a half parts each liquid propylene and liquid propane.

Light paths were employed almost three times as long as those which served for previous work in the visible region of the spectrum[1]. Dewars of special design were constructed for this work. Fused quartz windows were inserted by the use of graded pyrex-to-quartz seals in such a way that the graded seals were separated from direct contact with liquid nitrogen and thus from thermal shock by means of a gas space.

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[1] S. Freed and C. J. Hochanadel, J. Chem. Phys. In Press.

The spectra were unusually sharp, consisting of separated groups of lines. The lines from the gadolinium salt in solution were comparable in sharpness with those from the iron arc.

The spectrum of gadolinium lies in the ultraviolet in groups from 3100 Å to 2500 Å. (S. Freed, K. Sancier)

THE SOLUBILITY OF ARGON IN PROPANE AND ITS REACTION WITH BORON TRICHLORIDE DISSOLVED IN PROPANE AT 77° K

Propane (pure grade from Phillips Petroleum Corporation several times fractionated by distillation) was found to remain as a supercooled fluid for a half hour at 77° K. Upon circulating purified argon gas through the liquid at successively higher pressures, increased quantities of argon dissolved, forming an apparently stable fluid solution at 77° K. It was found that at about saturation one sixth mole of argon had dissolved for each mole of propane. Perhaps it should be recalled that the temperature is 6.6° below the freezing point of argon.

The viscosity of the argon solutions appeared less than that of pure propane which seems somewhat more than that of water at room temperature but roughly of the same order of magnitude. The surface tension was found to increase markedly after about one third the saturation concentration had been reached. Frothing set in then as bubbling of argon was going on.

Boron trichloride several times fractionated by distillation from commercial material about 94°/o pure, was distilled into propane at dry-ice temperature. The temperature was reduced, and the resulting solution remained fluid and homogeneous at 77° K. It contained about three mole per cent borontrichloride. This is about one sixth the concentration of argon in an approximately saturated **solution** in pure propane.

Purified argon gas measured out from a pipette was circulated through the solution of boron trichloride in propane at 77° K. When the solution had absorbed about one fiftieth the amount of argon corresponding to a saturated solution of argon in propane, the solution was observed to have become cloudy, and a precipitate began to settle out which grew in amount with every addition of argon. The rate of formation of the precipitate was rapid at the low temperature. The precipitate was flocculent at first, but settled out yielding a clear supernatant liquid. When the system was allowed to warm the precipitate disappeared at 123° K. However, when the temperature was again lowered to 77° K and more argon added, the resulting solid was found to dissolve completely at a lower temperature. For example, when the system had absorbed about one third as much argon as corresponds to the solubility in pure propane the system cleared at about 103° K, when about forty per cent as much argon had been added as corresponds to saturation in pure propane, the precipitate had "melted" into the solution at 100° K.

This set of experiments was originally undertaken with the view that the low temperature might favor the formation of compounds of argon and boron trichloride. High pressures had been employed in the work of Booth and Wilsson[1] in which they had found maxima in the phase diagram of argon-boron trifluoride (near its melting point, 143° K). These maxima indicated compounds of different molal ratios of argon to boron trifluoride in the solid state.

The successive lower temperatures at which our precipitates dissolved when increased amounts of argon were absorbed is consistent with the results of Booth and Wilsson if we assume that the composition of the precipitate is changing to higher molal ratios of argon to boron trichloride as more argon is

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[1] Booth and Wilsson, J.A.C.S. 57, 2273 (1935)

introduced. That is, the solubility is increasing; the heat of fusion presumably is decreasing.

It is intended to study compound formation in solutions at the low temperatures. (S. Freed, K. Sancier)

PROTACTINIUM CHEMISTRY

The purpose of the experiments which we have undertaken since the last progress report has been twofold; first, to isolate a small quantity of protactinium and second, to study certain aspects of the solution chemistry of this element. This report discusses experiments which have been carried out up to the present with a view toward achieving the first objective.

Protactinium occurs in all ores containing uranium, the amount present being about one half the quantity of radium present. When pitchblende is worked up commercially for uranium and radium some of the protactinium is carried along and ends in one of the several waste solids or solutions. We have investigated three of these wastes, which are designated as the "carbonates", "raffinates", and "filtered sparged turbid liquor", respectively. Shipments of these materials were arranged through the kindness of the Atomic Energy Commission and the Mallinkrodt Company.

Experiments with the "raffinates", a reddish-brown clay-like solid were disappointing in that our assays indicated that only a very small quantity of protactinium could be easily recovered. Following the lead of the Chicago (Argonne) group[1, 2] we turned to the "carbonates" and "filtered sparged turbid liquor" as more likely feed materials. Many experiments were performed on the "carbonates" in which the material was first attacked with any one of several acids; the dissolved phase (which carried no protactinium) discarded and the

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[1] Roy Thompson, AECD - 1897, February 1948

solid phase leached with hydrofluoric acid or fused with potassium bisulfate. Such procedures ended with disappointing results and seemed to offer little promise. The procedure described by Thompson[1] was then tried and somewhat stronger activities of protactinium were extracted. This procedure consists in treating the "carbonates" with a 40°/o sodium hydroxide solution heated to about 60° C. for four hours, followed by extraction of protactinium from the washed residue by means of nitric acid. We are at present engaged in working up a small quantity (about 300 grams) of the "carbonates" by this procedure.

The other waste material, "filtered sparged turbid liquor", a dark, acidic liquid which, belying its name, is actually fairly clear, has also been investigated. It was attempted to carry the protactinium from this liquid by means of a manganese dioxide precipitation but unfortunately on solution of the manganese dioxide precipitated from 50 gal. of the liquor a yellow precipitate appeared which again carried down all the protactinium. We are still engaged in working up this material. We are at present also studying two other procedures for the removal of Pa from the filtered liquor, the first of which, a solvent-extraction procedure, was suggested to us in a letter from D. F. Peppard at the Argonne National Laboratory[2]. The other involves carrying the protactinium on zirconium phosphate, with subsequent separation from the zirconium.

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[2] Letter: D. F. Peppard (Argonne Nat'l. Lab.) to Dr. R.W. Dodson, Jan. 3, 1950

Future plans include collaboration with the Reactor Hot Laboratory Operations group, in whose laboratory the necessary large-scale filtrations, extractions, etc., can be more conveniently carried out. It is hoped that milligram quantities or greater of Pa^{231} can be isolated and made available for research. (G. Harbottle)

NUCLEAR RECOIL FOLLOWING NEUTRINO EMISSION FROM Be^7

Some years ago Wang[1] suggested that the nuclear recoil from neutrino emission alone could be observed in the radioactive decay of K-capture nuclides, and that Be^7 was the best known nuclide for this purpose. More recently Crane[2] emphasized that it is important to show whether the nucleus emits one or more neutrinos following such a K-capture decay. This aspect of the problem is particularly important at the present time since it has been observed that the π-meson decays to a μ-meson and a single neutrino, while the μ-meson decays to an electron and two neutrinos. It is then possible that an experiment demonstrating whether K-capture decays are accompanied by single or multiple neutrino emission would furnish an important piece of information on the relation of electron-nucleon interaction with meson decay.

Single neutrino emission following K-capture in Be^7 would lead to a monoenergetic recoiling lithium atom of 57.3 e.v., and multiple neutrino emission would lead to a spectrum of recoil energies with an end point at 57.3 e.v. The technique employed in the present measurements was to distill carrier-free Be^7 from a tungsten ribbon on a surface, and to observe the ions recoiling from the surface with a vibrating reed electrometer after they have passed through a retarding grid system. All operations were carried out in a high vacuum system, and the sources moved inside the vacuum system magnetically. The tungsten ribbon was weighed on a microbalance before and after the distillation to measure the amount of contamination distilling along with the Be^7 (the weight of Be^7 itself is far too low to measure). From a knowledge of

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[1] K. C. Wang, Phys. Rev. 61, 97 (1942)

[2] H. R. Crane, Rev. Mod. Phys. 20, 278 (1948)

the geometry of distillation an estimate of the upper limit of the weight of the deposit can be obtained. Weights so obtained for the various surfaces studied are listed in Table I. At the conclusion of each experiment the intensity of the source was measured by a comparison of its gamma counting rate with that of a Co^{60} standard (10 percent of the K-capture decays of Be^7 go to an excited state of Li^7 and are followed by emission of 0.480 Mev gamma rays[3]). From the source strength an estimate can be made of the total number of recoils expected, and this is compared to the measured ion current. The yields of positive ions and electrons computed in this way are listed in Table I for several different kinds of surfaces.

Table I

	Positive Ions			Negative Ions		
Surface	Weight of **Deposit**	i_+, current amps X 10^{16}	Percent Yield	i_-, current amps X 10^{16}	Percent Yield	i_+/i_-
Beryllium (freshly distilled)	$<3\pm1 \times 10^{-6}$	1	3	28	70	0.04
Platinum foil	---	7	4	33	20	0.21
Platinum (outgassed 950° for 60 hrs.)	$2\pm1 \times 10^{-6}$	6	20	19	60	0.31
Tungsten foil	---	20	--	120	--	0.16
Mica, freshly cleaved	$<3\pm1 \times 10^{-6}$	7	20	1.4	4	5.00

From the data it is clear that the ratio of positive ions to electrons is much higher for a mica surface than for any of the metal surfaces used. In general a low yield of ions would be expected from surfaces having a work function lower than the ionization potential of lithium (5.36 e.v.). Although

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[3] C. M. Turner, Phys. Rev. 76, 148 (1949)

it is difficult to say quantitatively what the work functions of the above surfaces are, particularly in the neighborhood of a recoiling atom, one would expect a freshly deposited beryllium surface to have a work function a volt or two below the ionization potential of lithium. Further, one would expect mica to exhibit a high work function, and therefore a large yield of positive ions is to be anticipated with a mica surface. The data of Table I are in qualitative agreement with these ideas. The best measurements of the ion energies by use of the retarding grid were obtained for the mica surface because in this case the ratio i_+/i_- was largest. The observed retardation curve (Fig. 1) does not drop off abruptly, as would be required if single neutrino emission occurred, and one concludes that the energy of the recoiling ions from these sources varies in energy from some very low value up to a maximum of around 30 to 40 e.v. The exact end point cannot be determined accurately because of the gradual approach of the curve to zero, and because of a lack of knowledge of the contact potential between the source and the grid system. The gradual falling off observed can be explained by the loss of energy of the recoiling atoms through collisions with contaminants on the surface. All similar retarding potential curves observed for Be^7 have exhibited similar behavior [4, 5]. However the latest work referred to above reports that ions of energies up to 56 e.v. were observed. In order to measure the recoil spectrum accurately enough to distinguish definitely between single and multiple neutrino emission one must prepare surfaces clean enough so that a nearly 100 percent yield of recoiling atoms is obtained.

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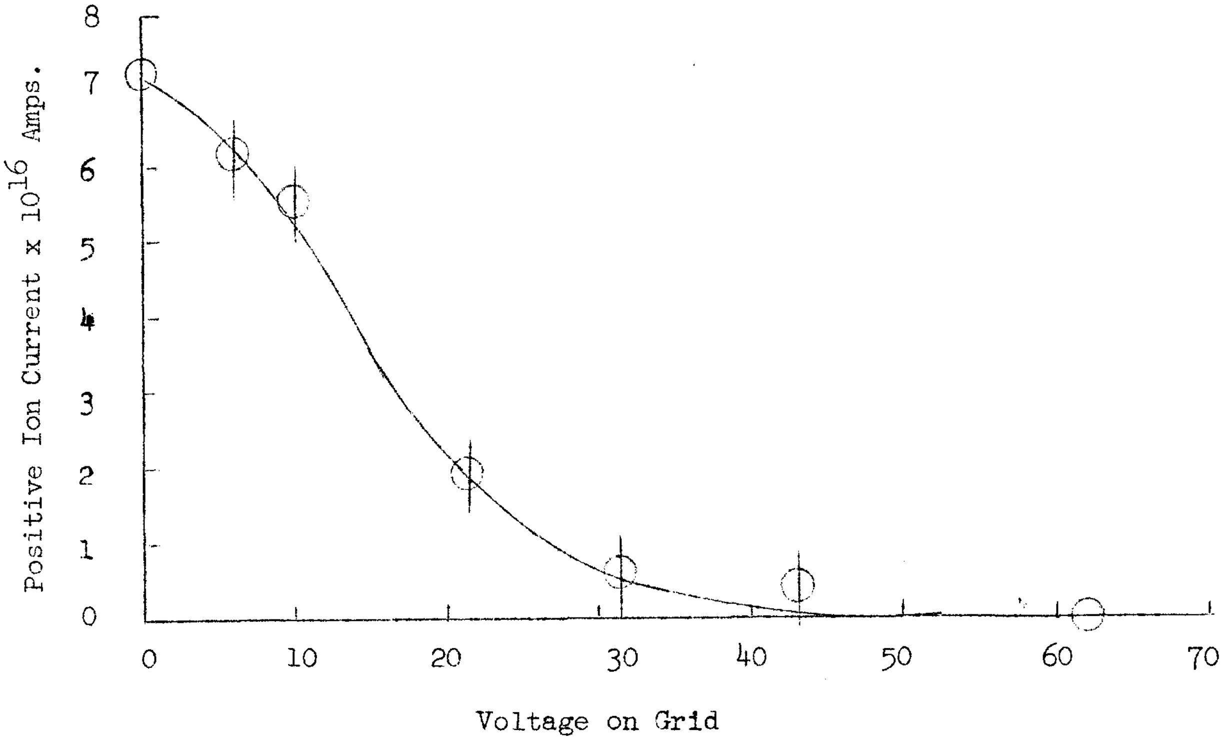
[4] J. S. Allen, Phys. Rev. 61, 692 (1942)

[5] P. B. Smith and J. S. Allen, Bull. Am. Phys. Soc. 24 No. 7, 14 (1949)

Fig. 1



Two pieces of apparatus are being prepared to continue this study. In one the energy of the recoiling ions will be measured with a potential grid system and an electrometer, and it will be possible in this apparatus to prepare surfaces about one hundred times cleaner than in the above experiment (about 10^{-8} gms/cm^2). This apparatus has been completed, and tried out, but unfortunately the Be^7 source available at the time was not active enough to allow measurements. The other apparatus is an electrostatic analyser with an electron multiplier as a detector. The electron multiplier is now being tested, otherwise the apparatus is complete.

(Raymond Davis, Jr.)

THE THALLOUS-THALLIC EXCHANGE REACTION

As reported in the previous progress report, the electron transfer exchange reaction between thallous and thallic thallium is being studied further in the chloride system where the rate of this reaction is greatly affected by the chloride ion. From the data obtained to date in two series of runs at different ionic strengths ($\mu = 0.5$ and $\mu = 3.0$) and with chloride concentrations ranging from zero to one formal, it is apparent that the rate function (over-all bimolecular rate constant) passes through a minimum as the chloride concentration is increased, the minimum occurring at low chloride concentrations. This deceleration of the rate of exchange at low chloride concentrations may be attributed to the formation of a chloride complex of one of the reacting species which exchanges at a slower rate than the species present when there is no chloride present. Then after sufficient chloride has been added a higher chloride complex may be formed which exchanges rapidly.

A comparison of the curves obtained when the rate function is plotted against the chloride concentration for $\mu = 0.5$, 3.0 and $\mu = 6$ (from earlier work done by G. Harbottle, Thesis, Columbia University, 1949) indicates that there is little, if any, ionic strength effect as the chloride concentration is varied from 1f down to approximately 0.1f. However, at chloride concentrations lower than about 0.1f the curves start to fan out, with the rate function decreasing as the ionic strength increases.

Experiments done at a chloride concentration of 0.033f have shown that the use of either sodium perchlorate or perchloric acid to adjust the ionic strength gives the same rate of exchange. An experiment, also performed at a chloride concentration of 0.033f, in which the ionic strength was doubled from 0.0343 to 0.0686 by the addition of sodium perchlorate, resulted in essentially the same rate function for both ionic strengths.

It is hoped that continuation of these measurements will yield data which will permit characterization of the reacting species at various chloride concentrations, and that the rate data can be correlated with equilibrium measurements on the stability of the several chlorothallate complex ions. (L. Eimer and R. W. Dodson)

THE EXCHANGE REACTION BETWEEN FERROUS AND FERRIC IONS IN PERCHLORIC ACID SOLUTIONS

It has been variously reported that the electron transfer exchange reaction between ferrous and ferric ions is immeasurably fast[1], very slow[2], or complete in one to two hours[3]. Some of the discrepancies in past work have been attributed to catalysis of the exchange reaction during the chemical separations employed to separate the exchanging species. However, Linnenbom and Wahl[3], using a diffusion separation were unable to confirm the result of Van Alten and Rice[2], who reported the reaction to require weeks for completion when separations were performed in this way. A study of the reaction has been undertaken with the hope of resolving these discrepancies, and to round out the work on other electron transfer reactions (cerous-ceric, and thallous-thallic) being carried out in this laboratory. A quantitative chemical separation has been devised which leads to only a small amount of "induced exchange". The results which have been obtained to date indicate that the reaction is indeed a rapid one but that its rate can be measured.

The separation method is based on complexing the ferrous ion with α-α'- dipyridyl. The resulting complex ion is stable over a wide range of

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[1] P. Nahinsky, Ph.D. Thesis, Univ. of Calif.,(1942)

[2] L. Van Alten and C. N. Rice, J. Am. Chem. Soc. 70, 883 (1948)

[3] V. J. Linnenbom and A. C. Wahl, J. Am. Chem. Soc. 71, 2589 (1949)

pH, and even in solutions in which it is unstable decomposes only slowly. After the complex is formed the ferric species can be separated by any of several common precipitation methods; we have used precipitation as ferric hydroxide.

The procedure employed is to mix the reactants with rapid and continued stirring, and at the end of the selected reaction time to add in rapid succession an excess of a dilute solution of dipyridyl followed by sufficient sodium acetate solution to bring the pH to about 5. Subsequently the ferric iron is precipitated by adding ammonium hydroxide. The iron in either or both fractions is finally converted to ferric 8-hydroxyquinolate and counted as such. The tracer employed in the experiments here reported was Fe^{55}, obtained from Oak Ridge.

With this procedure the zero time exchange ranges from 15 to 30°/o. The fraction of complete exchange exhibits the usual exponential time dependence with a half-time of about 20 seconds, under the conditions of the experiments. It is believed that catalysis by traces of chloride is negligible, in view of the data in the following table, which summarizes the results so far obtained.

$HClO_4$ formal	Fe^{+++} formal	Fe^{++} formal	Chloride	Half-Time
0.4	0.83×10^{-3}	1.06×10^{-3}	trace (ca 10^{-3})	23±2 seconds
0.4	0.97×10^{-3}	1.02×10^{-3}	none detectable (10^{-6} or less)	20±2 "
0.4	"	"	" "	18±3 "
0.4	"	"	0.8×10^{-3}f added	21±2 "

All runs at room temperature 23±2° C.

The kinetics of the reaction are being further investigated with respect to: reaction order, temperature coefficient, acid and salt effects. (R.W. Dodson)

ISOTOPE EFFECT ON BOND RUPTURE BY ELECTRON IMPACT ON HYDROGEN, DEUTERIUM AND TRITIUM

There have been several experimental and theoretical estimates of the relative yields by electron impact of atomic and molecular ions for hydrogen and deuterium[1,2,3]. The previous experimental results were obtained by mass spectrometric measurements using low velocity electrons.

Although the ratio of the ratios H^+/H_2^+ and D^+/D_2^+, as reported, was in satisfactory agreement with the predicted value, the discrepancy in the individual ratios, X^+/X_2^+ was large. It was thought that this discrepancy was caused mainly by "voltage discrimination", as defined by Coggeshall[4], in the mass spectrometer source. An investigation of this point together with the availability of tritium prompted this research.

A 60° sector type mass spectrometer was used. The two important features of the spectrometer as far as present work was concerned, were an electronically regulated 5000-volt accelerating voltage and a vibrating reed electrometer providing extremely sensitive detection. The 5000-volt supply enabled us to investigate the effect of voltage discrimination; a summary of the results is given in Table I.

TABLE I

Relative Ion Yields as a Function of Accelerating Voltage

Acc. Voltage	H^+/H_2^+	D^+/D_2^+	T^+/T_2^+
2500	1.1%	0.57%	0.31%
3000	1.2	0.62	0.31
3500	1.3	0.67	0.35
4200	1.3	0.70	0.36

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[1] W. Bleakney, E. U. Condon and L. G. Smith, J. Phys. Chem. 41, 197 (1937)

[2] N. Bauer and J. Y. Beach, J. Chem. Phys. 15, 150 (1947)

[3] D. Stevenson, J. Chem. Phys. 15, 409 (1947)

[4] N. Coggeshall, J. Chem. Phys. 12, 19 (1944)

These results are believed to indicate that voltage discrimination is of small consequence above 4000 volts in the ion source employed.

The theoretical calculations of Stevenson[3] were based upon the assumption that the excitation by electron impact could be considered as a Franck-Condon type transition. We have extended these calculations to the case of tritium. The comparison of calculated values with the previously reported experimental observations and with those of the present work is given in Table II.

TABLE II

Relative Ion Yields Predicted and Observed

	H^+/H_2^+	D^+/D_2^+	T^+/T_2^+	$\frac{H^+/H_2^+}{D^+/D_2^+}$	$\frac{H^+/H_2^+}{T^+/T_2^+}$	$\frac{D^+/D_2^+}{T^+/T_2^+}$
Anharmonic Oscillator Calculated Value	1.87°/o	0.83°/o	0.44°/o	2.3	4.3	1.9
This research, 30 volt electrons	1.3	0.70	0.36	1.9	3.7	1.9
Hipple[1], 22 volt electrons	0.96	0.34		2.8		
Bauer and Beach[2], 30 volt electrons	0.58	0.38		1.5		

It is seen that there is much closer agreement between calculated and experimental values than was previously found. We have observed that the positioning of the source magnet, because of its effect on the focussing properties of the ion source, has an influence on the ratios. This effect is most pronounced for the H^+/H_2^+ ratio since the magnetic fields of the source and scanning magnets are comparable. This accounts for the relatively poorer agreement for this ratio. We believe that the previously observed discrepancies were due, in the main, to voltage discrimination. The remaining differences are larger than can be explained by errors in the experimental

measurements. However, the approximation to the wave function and the assumption that the ionization probabilities are voltage independent make the calculated individual ratios too large. The results obtained demonstrate that the electron impact excitation can indeed be considered as similar to the optical case and treated by means of the Franck-Condon principle.
(O. Schaeffer, J. Hastings.)

OXYGEN AND NITROGEN ISOTOPE EFFECTS IN THE DECOMPOSITION OF AMMONIUM NITRATE

The decomposition of ammonium nitrate offers the possibility of investigating both nitrogen and oxygen isotope effects in a non-equilibrium system; one can start with enriched nitrogen in either the ammonium ion or the nitrate ion and determine whether the N_2O formed is predominantly $N^{14}N^{15}O$, $N^{15}N^{14}O$ or a mixture of these; an isotopic assay of the oxygen in both the N_2O and the water formed provides a possible method for the analysis of the isotopic composition of the oxygen in nitrates; in the event that either of the isotopic isomers $N^{15}N^{14}O$ or $N^{14}N^{15}O$ is produced exclusively the infrared spectrum of the isomer can be determined. This would provide a basis for the improvement of the force constants and potential function for the vibrations of N_2O.

Decomposition of $N^{15}H_4NO_3$

In a preliminary attempt to prepare N_2O from NH_4NO_3 a 3 gram sample of Baker's "Analyzed" NH_4NO_3 was put in a tube connected to a vacuum system. The tube was surrounded by a furnace and the sample was pumped at approximately 150° C over night. While NH_4NO_3 sublimes freely in vacuo, no decomposition was observed up to 300° C. Addition of a few cm. pressure of tank N_2O sufficed to prevent sublimation, but did not catalyze the decomposition. On the other hand, addition of small amounts of water vapor to a dried and degassed sample induced the decomposition when the temperature was raised to 200° C. The reaction was carried to completion. During the course of the decomposition the water was continuously removed from the gas stream. The gas sample contained over 98°/o N_2O and the remainder was N_2 and O_2. The N_2 and O_2 which were formed in reproducible amount come either from some side reaction or the decomposition of N_2O in the hot portion of the apparatus.

A sample of N_2O was prepared from NH_4NO_3 containing excess N^{15} in the $NH_4{}^+$ ion. The NH_4NO_3 contained nominally 7.5°/o excess N^{15} and was obtained from the Eastman Kodak Company. A 2.4 g sample was decomposed after thorough evacuation and drying and then the addition of trace amounts of water. The N_2O was collected and analyzed mass spectrometrically in a Consolidated-Nier Isotope Ratio mass spectrometer. The results of the analyses are given in Table I.

TABLE I

Peak Measured	Ion	Ratio
45/44	N_2O^+	0.0869
31/30	NO^+	0.010
29/28	$N_2{}^+$	0.081

If the isomer $N^{15}N^{14}O$ is produced then the intensity ratio of 31/30 corresponding to the $N^{15}O^+/N^{14}O^+$ ions should be that of normal N^{15} isotopic abundance or 0.0044. On the other hand if the isomer $N^{14}N^{15}O$ is produced then this ratio should be equal to the 45/44 ratio of molecular ions. If any combination of these is produced the 31/30 ratio will be between these limiting values. The possibility of rearrangement on electron impact in the mass spectrometer may give a value intermediate between 0.004 and the 45/44 ratio even if an authentic sample of $N^{15}N^{14}O$ were analyzed. However, fortunately such rearrangements appear to be uncommon and where they do occur are usually less than 10°/o of the more normal simple rupture of chemical bonds by electron impact. It is clear from the data in Table I that our sample contains less than 7°/₀ of the isomer $N^{14}N^{15}O$. The excess of the 31/30 ratio observed may be due to either production of $N^{14}N^{15}O$ in the course of the decomposition reaction or to production of $N^{15}O^+$ ions from $N^{15}N^{14}O$ by electron impact or a combination of both. Therefore, the 7°/₀ $N^{14}N^{15}O$ from NH_4NO_3 containing N^{15} in the NH_4^+ ion is an upper limit.

If one carries over some of the accepted ideas[1,2,3] of the oxidation of NH_4^+ as well as the reduction of NO_3^- ion in aqueous solution to the reaction occurring in the molten NH_4NO_3 one might consider the following sequence of reactions leading to the products:

$$\begin{array}{lll}
NH_4^+ + NO_3^- \longrightarrow NH_3OH^+ + NO_2^- & & (1) \\
NH_3OH^+ + NO_2^- \longrightarrow 2NOH + H_2O & & (2a) \\
NH_3OH^+ + NO_2^- \longrightarrow H_2N_2O_2 + H_2O & & (2b) \\
2NOH \longrightarrow N_2O + H_2O & & (3a) \\
H_2N_2O_2 \longrightarrow N_2O + H_2O & & (3b)
\end{array}$$

It is clear that the sequence of reactions which produces N_2O through nitroxyl would lead to equal amounts of the isomers $N^{15}N^{14}O$ and $N^{14}N^{15}O$, apart from any small isotopic fractionation in the process. This mechanism is ruled out conclusively by the results in Table I. The sequence of reactions involving hyponitrous acid is similarly ruled out unless one postulates an unsymmetrical structure for hyponitrous acid. Most of the present evidence points to the

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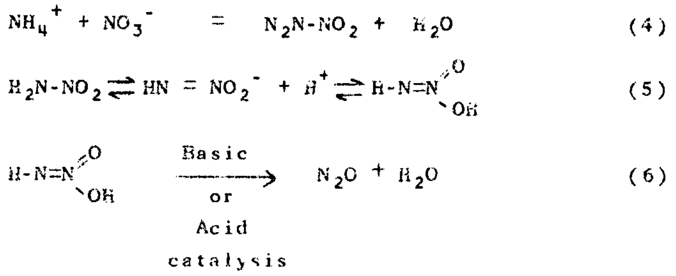
[1] L. H. Milligan and G. R. Gillette, J. Phys. Chem. 28, 746 (1924)

[2] E. Abel and J. Proisl, Monatshefte Für Chemie, 66, 6 (1935); 70, 201 (1937); 72, 1 (1938)

[3] W. C. Bray and G. K. Rollefson, "Manual of Advanced Inorganic Chemistry", Printed Privately, July 1941.

structure HONNOH for this acid[4,5].

Alternatively one may consider the production of N_2O from NH_4NO_3 through nitramide as an intermediate. A possible sequence of reactions is:



$$NH_4^+ + NO_3^- = N_2N\text{-}NO_2 + H_2O \qquad (4)$$

$$H_2N\text{-}NO_2 \rightleftharpoons HN = NO_2^- + H^+ \rightleftharpoons H\text{-}N{=}N\begin{matrix}{\nearrow}O \\ {\searrow}OH\end{matrix} \qquad (5)$$

$$H\text{-}N{=}N\begin{matrix}{\nearrow}O \\ {\searrow}OH\end{matrix} \xrightarrow[\substack{\text{or} \\ \text{Acid} \\ \text{catalysis}}]{\text{Basic}} N_2O + H_2O \qquad (6)$$

Steps 5 and 6 are the mechanisms proposed by Marlies and La Mer[6] and Pederson[7] to explain the acidic and basic catalysis of nitramide decomposition.

Reaction Initiated by H_2O^{18}

An experiment was carried out to determine whether the water catalysis was primarily chemical or physical in nature. In one experiment 4.6 c.c. of water vapor containing 1.4 °/o O^{18} was used to initiate the decomposition of a three gram sample of ordinary NH_4NO_3, which had been previously thoroughly dried and outgassed. The decomposition was allowed to progress until about 4 c.c. of N_2O were produced. The O^{18} content of the N_2O was within 0.5 °/o of that of N_2O formed under similar conditions but with the reaction initiated by ordinary water. There is obviously no exchange between N_2O and H_2O under these experimental conditions. If the water containing enriched O^{18} had exchanged with all of the oxygen in NH_4NO_3 then the N_2O should be enriched in O^{18} over the sample obtained from the ordinary water reaction by 1.6 °/o. Such

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[4] Hunter and Partington, J. Chem. Soc. 1933, 309
[5] A. Hantzsch, Ber. 66, 1555 (1933)

[6] C. A. Marlies and V. K. La Mer, J. Am. Chem. Soc. 57, 1812 (1935)
[7] K. J. Pederson, J. Phys. Chem. 38, 581 (1934)

an effect would have been easily determined and the water catalysis cannot be explained in this way. At present we see no explanation for the water catalysis.

In a separate experiment N_2O was allowed to stand over water containing 1.4°/o O^{18} for three days at 24 ± 1° C. Analysis of the N_2O for O^{18} showed 0.26°/o exchange. This is an upper limit for the exchange for the water may have contained a small amount of CO_2 which would have come to equilibrium in this time. Both CO_2 and N_2O have the same molecular weights and both would be measured simultaneously in the mass spectrometer. The exchange will be studied in CO_2-free water as a function of temperature and pH. It is not surprising that N_2O does not exchange with water. Solutions of N_2O in water show no acidic properties[8] despite the fact that N_2O is the stoichiometric anhydride of hyponitrous acid. Furthermore N_2O is salted out of water by all electrolytes, including bases, except nitric acid[8,9].

Isotope Effects in the Decomposition of NH_4NO_3 and the Isotopic Composition of NH_4NO_3.

A study of the isotope effects in the decomposition of ammonium nitrate was undertaken to investigate the magnitude of these effects in the rupture of nitrogen-oxygen bonds and perhaps to aid in the elucidation of the mechanism of the reaction. At present the nitrogen isotope effects are being studied with both normal and enriched N^{15}. The oxygen isotope effects were studied with normal O^{18} and at present provisional values can be given. Two

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[8] G. Geffcken, Z. physik. Chem. 49, 301 (1904)

[9] Lunge, Ber. 14, 2118 (1881)
Knop, Ibid., 48, 106 (1904)
Manchot, Z. Anorg. Chem. 141, 38 (1924)

separate determinations have been made. One was in a reaction initiated by normal water, while the other was initiated by water containing enriched O^{18}. While none of the O^{18} from the enriched water appeared in the N_2O after a small amount of decomposition, it cannot be stated that the fate of the enriched water after complete reaction is simply dilution with the water from the decomposition. Using the latter assumption we find the results of the fractionation study when enriched water was used consistent with that using ordinary water. We shall report only the results of an experiment in which the decomposition was initiated by the addition of a minute amount of ordinary water.

The reaction was carried out in the method described previously. The decomposition temperature was 220 ± 20° C. Temperature control was difficult because of the large heat evolved in the reaction. The N_2O formed after 1°/o decomposition was collected in a bulb. The reaction was then carried to completion and the N_2O and water were separated by cold traps. The N_2O was freed of the small amounts of nitrogen and oxygen and analyzed as N_2O. The water was equilibrated with CO_2 and analyzed according to the method described by Cohn and Urey[10]. It does not seem feasible to obtain the water at 1°/o decomposition. Each sample was analyzed at least three times and the reproducibility of the analysis was better than 0.3°/o. The results are given in Table II.

TABLE II

O^{18} Analyses of N_2O and H_2O

Sample	46/44+45 Ratio
Tank CO_2	0.004183
CO_2 from H_2O equilibration	0.004285
N_2O $t = \infty$	0.002127
N_2O $t = \epsilon$	0.002117

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[10] M. Cohn and H. C. Urey, J. Am. Chem. Soc. 60, 679 (1938)

To determine that 46/44 ratio and the mole ratio of O^{18}/O^{16} the 45/44 ratios were determined for CO_2 and N_2O. Under the same operating conditions of the spectrometer they were 0.0122 and 0.00798 respectively. None of the ratios should be considered absolute determinations of O^{18}. Fractionation in the ion source and leak and errors in the amplifier intercalibrations cancel out in the relative determinations because of the fact that both gases are measured for O^{18} by a 46/44+45 ratio. The 45/44 ratio introduces a small correction. The data in Table II lead to the provisional fractionation factors listed in Table III.

TABLE III

Fractionation Factors

$\left(\dfrac{N_2O^{18}}{N_2O^{16}}\right)_{t=\infty} \Big/ \left(\dfrac{N_2O^{18}}{N_2O^{16}}\right)$	$t = \epsilon$	1.005 ± .003
$\left(\dfrac{N_2O^{18}}{N_2O^{16}}\right)_{t\infty} \Big/ \left(\dfrac{H_2O^{18}}{H_2O^{16}}\right)$	$t = \infty$	1.018 ± .003
$\left(\dfrac{NH_4NO_2{}^{16}O^{18}}{NH_4NO_3{}^{16}}\right) \Big/ \left(\dfrac{H_2O^{18}}{H_2O^{16}}\right)$	ordinary water	3 × (1.030 ± 0.003).

The conversion of the $CO^{16}O^{18}/CO_2^{16}$ to the H_2O^{18}/H_2O^{16} ratio was made using the equilibrium constant for the reaction

$$CO_2^{16} + H_2O^{18}(l) = CO^{16}O^{18} + H_2O^{16}(l) \qquad (7)$$

$$K_7 = 2 \times 1.037 \text{ at } 24^\circ \text{ C.}$$ [10, 11]

The isotopic analysis for the NH_4NO_3 was calculated from the N_2O^{18}/N_2O^{16} and H_2O^{18}/H_2O^{16} ratios and the stoichiometry of the reaction

$$NH_4NO_3 = N_2O + 2H_2O \qquad (8)$$

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[11] H.C. Urey, J. Chem. Soc. 1947, 562

To obtain the ratio $\left(\frac{NH_4NO_2{}^{16}O^{18}}{NH_4NO_3{}^{16}}\right) \Big/ \left(\frac{H_2O^{18}}{H_2O^{16}}\right)$ ordinary water

the CO_2 to H_2O conversion factor K_7 above was used. This is justified by the observations that tank CO_2 is in equilibrium with ordinary water with respect to oxygen exchange at room temperature[10]. The temperature coefficient of (7) is small at room temperature[11].

From the fact that the vibrational spectrum of nitrate ion is extremely similar to that of carbonate ion[12] and the fact that carbon and nitrogen are not too different in mass one would expect the oxygen isotopic composition of nitrates in equilibrium with water to be very similar to that of carbonates in equilibrium with water. From the analysis of limestones it is found[13] that

$\left(\frac{CO_2{}^{16}O^{18=}}{CO_3{}^{16=}}\right) \Big/ \left(\frac{H_2O^{18}}{H_2O^{16}}\right)$ is equal to 3 x 1.035. We are investigating Ordinary water

the source of commercial nitrates to ascertain the probability that these nitrates are in equilibrium with water. The above method seems to be a good method for the isotopic analysis of oxygen in nitrates.

The fractionation factor of oxygen in N_2O during the course of the reaction is small as is to be expected for reactions occurring at 500° K. This ratio depends only on the zero point energy effect[14,15]. The zero point energy

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[12] G. Herzberg, Infra-Red and Raman Spectra of Polyatomic Molecules, D. VanNostrand and Co., 1945, pg. 178

[13] B. F. Murphey and A.O. Nier, Phys. Rev. 59, 771 (1941)

[14] J. Bigeleisen, J. Chem. Phys. 17, 675 (1949)

[15] J. Bigeleisen, Science, 110, 14 (1949)

effects at 500° K. are at most five per cent[11]. The fractionation factor of

$$\left(\frac{N_2O^{18}}{N_2O^{16}}\right) \Big/ \left(\frac{H_2O^{18}}{H_2O^{16}}\right)_{t=\infty}$$ depends on the mechanism of the reaction.

In view of the fact that the mechanism is not clearly established all we wish to state at present is that the observed factor is consistent with the rupture of a nitrogen-oxygen bond where the symmetry of nitrate ion is destroyed. If exchange equilibria between various intermediates and water and N_2O were established one would expect a fractionation factor of no more than about two per cent. Detailed calculations of some of the exchange equilibria are being made and additional samples are being prepared for analysis.

Vibrational Spectrum and Thermodynamic Properties of the Isotopic N_2O Molecules.

Both the infra-red[16] and Raman[17] spectra of N_2O show conclusively that N_2O is a linear unsymmetrical molecule. The observed frequencies lead to complex force constants in the valence force potential function for a linear XYZ molecule. In order to improve the potential function and to obtain the vibrational spectra of the isotopic N_2O molecules necessary for the calculation of the thermodynamic properties[18] the infra-red spectrum of 8 °/₀ $N^{15}N^{14}O$ was determined. All the fundamental frequencies of all of the isotopic N_2O molecules occur in the experimentally accessible infra-red region.

The spectrum of $N^{15}N^{14}O$ was determined from a differential spectrum of 8% $N^{15}N^{14}O$ in $N^{14}N^{14}O$ vs. $N^{14}N^{14}O$. The measurements were made in a Baird Associates double-beam recording infra-red spectrophotomer, with both NaCl

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[16] E. K. Plyler and E. F. Barker, Phys. Rev. 38, 1827 (1931)
[17] A. Langseth and J. R. Nielson, Nature, 130, 92 (1932)

[18] J. Bigeleisen and M. G. Mayer, J. Chem. Phys, 15, 261 (1947)

and KBr optics. Measurements were made at different total pressures, but always with the same total pressure in each cell. This procedure gave a spectrum with a negative absorption for $N^{14}N^{14}O$ and a positive absorption for $N^{15}N^{14}O$. The wave length separations measured both manually and automatically were converted to wave number separations by means of the relation $\Delta\nu=-\bar{\nu}^2\Delta\lambda$. The procedure was checked using the separation of the doublets in the NH_3 spectrum in the same spectral ranges. The agreement with Barker's[19] measurements of the separation of the NH_3 doublets was within ±2 cm^{-1}. In the case of the 2223 cm^{-1} band the P branch of the $N^{14}N^{14}O$ overlapped the R branch of the $N^{15}N^{14}O$ band. In the 1285 cm^{-1} band they were clearly resolved and separated. The shifts in the 2223.5 cm^{-1} region were obtained by subtraction of the P-R separation from the distance between $R(N^{14}N^{14}O)$ and $P(N^{15}N^{14}O)$. We also noticed a shift in the 3481 cm^{-1} band but no measurements were made because of the poor resolution of NaCl optics in this region. The shift in the 588.8 cm^{-1} band was easily resolved and determined from the separation of Q branches. The shifts are reported in Table IV

TABLE IV

Band	$\Delta\nu$ obs.
588.8	4.2 ± 0.4
1285.	16.1 ± 2
2223.5	29. ± 4

Knowledge of these shifts and application of the product rule allows a comparison of the observed and calculated shifts for the two symmetry classes of the $C_{\infty v}$ molecule.

Ratio	Observed	Calculated
$\frac{\omega_2}{\omega_2'}$	1.007	1.006
$\frac{\omega_1\omega_3}{\omega_1'\omega_3'}$	1.026	1.024

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[19] E. F. Barker, Phys. Rev. 55, 657 (1939)

It is seen that the product rule is checked within the accuracy of our experimental measurements. The frequencies as well as the frequency shifts will be used to evaluate the three force constants in the most general potential function for a linear triatomic molecule.

After our experiments on the infra-red spectrum were completed we were informed that a similar infra-red study was made independently by Richardson and Wilson[20]. Richardson and Wilson have measured the shifts only in the 2223.5 and 1285 cm^{-1} bands using 60 per cent $N^{15}N^{14}O$. Their value in the 1285 band is in good agreement with ours. They measured a shift of 22.5 cm^{-1} at 2223.5 cm^{-1} in contrast to our value of 29 ± 4 cm^{-1} obtained by the differential technique. The difference is just outside the estimated experimental error.

After a new potential function is evaluated the thermodynamic properties of all of the isotopic N_2O molecules will be calculated. This will permit the first reliable calculation of the thermodynamic equilibria involving isotopic isomers.

We wish to thank Mr. W. S. Richardson and Professor E. B. Wilson, Jr. for lending us a manuscript of their paper in advance of publication.
(L. Friedman, J. Bigeleisen)

THE SYNTHESIS OF L-ASCORBIC ACID UNIFORMLY LABELED WITH C^{14}

The synthesis of L-ascorbic acids isotopically labeled in different positions is of great interest in biochemical research. Recently Burns and King[1] have described a synthesis of 1-C^{14}-L-ascorbic acid*. We wish

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[20] W. S. Richardson and E. B. Wilson, Jr., J. Chem. Phys. In press.

[1] Burns and King, Science, 1950, 111, 257

to report the synthesis of uniformly enriched L-ascorbic acid*. It is worth noting that the uniformly labeled and 1-labeled compounds may play complementary roles in tracer experiments with this compound. Our synthesis was patterned after that of Reichstein and Grüssner[2] adapted to working on a semi-micro scale.

A mixture of uniformly enriched sucrose, glucose, and fructose was prepared by allowing bean leaves to photosynthesize for 24 hours in an atmosphere of $C^{14}O_2$[3]. The sucrose was hydrolyzed and a mixture consisting mainly of glucose was precipitated by the addition of absolute ethanol. The mixture, weighing 392 mg and having a specific activity of 1.4 μc/mg, was hydrogenated in an alkaline aqueous medium using Raney nickel. The sorbitol produced was oxidized to sorbose with Acetobacter suboxydans by the method of Wells, Stubbs, Lockwood, and Roe[4]. After the fermentation the organism was removed by centrifuging, and the supernatant liquid was passed through amberlite 1R-100-H and Duolite A-4 ion exchange columns to remove ionic impurities. Crystalization and decolorization with Darco G-60 gave 344 mg of pure sorbose, m. 163-5°.

The sorbose was diluted with 135 mg of carrier, and acetonated, using freshly distilled dry acetone, and sulfuric acid as a catalyst. The di-

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[2] Reichstein and Grüssner, Helv. Chim. Acta, 1934, 17, 311

[3] M. Gibbs, "Biochemical Preparations", New York, John Wiley and Sons, in press

[4] Wells, Stubbs, Lockwood, and Roe, Ind. and Eng. Chem., 1937, 29, 1385

* According to suggestions for the naming of isotopically labeled compounds proposed at the 4th Chemistry Conference, Brookhaven National Laboratory, and shortly to be published, these acids would be known as L-ascorbic acid-1-C^{14}_{1}, and L-ascorbic acid-ue-C $^{14}_{6}$, respectively.

acetonesorbose was extracted from unreacted material and monoacetone derivative with ether. Oxidation of the diacetonescorbose with 6 per cent potassium permanganate was carried out in alkaline medium[5], the unattacked material from the first oxidation being removed by ether extraction and reoxidized. The two batches were carried through separately to the last step. The reaction mixture was filtered and acidified to pH2 after which the diacetone-2-ketogulonic acid was extracted with ethyl acetate and refluxed with water for 40 minutes to effect deacetonation. Lyophilization gave white, powdery 2-ketogulonic acid, m. 170-1°. The methyl ester was prepared by passing a large excess of diazomethane into a methanolic solution of the acid at -15°C. By crystallization from methanol-acetone, 68 mg of white crystals, m. 145-50° was obtained. There was obtained in addition, 220 mg of residual oil, consisting partly of methyl 2-ketogulonate, which could not be induced to crystallize, and was therefore diluted with 205 mg of carrier, and carried through the last step. Treatment under nitrogen of the methanolic solutions of the ester with stoichiometric amounts of 4.65N sodium methoxide in methanol and 2.60N hydrogen chloride in methanol, gave, after purification according to the method of Szent-Gyorgyi[6], 30 mg of L-ascorbic acid having specific activity of 0.80 μc/mg and 70 mg having specific activity 0.16 μc/mg. The products melted at 189-190°, and gave a single spot of proper R_f value on a paper chromatogram.

The sample so prepared will be used by the Brookhaven Biology staff for plant and animal experiments on the metabolism of this ascorbic acid. (A. Bothner-By and R. C. Anderson, in collaboration with M. Gibbs of Biology)

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[5] Maksimov, J. Gen. Chem. (U.S.S.R.), 1939, 9, 936

[6] Szent-Gyorgyi, Biochem. J., 1928, 22, 1394

THE DECAY SCHEME OF Ni 57

Pulse analyzers and proportional counters suitable for the detection and identification of X-rays[1,2] have recently been developed. With the aid of such an instrument, the quantitative investigation of electron capture processes is being undertaken. The first case studied is that of 36-hour Ni^{57}, which, it has been reported, emits 0.67-0.72-Mev. positrons, 1.97-Mev gamma rays, and some softer gamma rays[3,4], but which we had reason to suspect of decaying by electron capture also.

The Ni^{57} was produced by bombardment of iron with 30-Mev alpha particles in the cyclotron of the Department of Terrestrial Magnetism of the Carnegie Institution. With the aid of carriers the nickel activity was chemically separated from the target and purified from other activities - especially those of cobalt, manganese, and copper - which may have been produced by alpha particles, deuterons, or neutrons either in the iron or in its supporting materials. The treatment involved dissolution of the iron target in 12N HCl; oxidation of the iron to the ferric state and its removal from the solution by ether extractions; and precipitation then of mickel dimethylglyoxime in the presence of tartrate ion to keep ferric ion in solution and of carriers to "hold back" cobalt, manganese, and copper. The nickel precipitate was dissolved and reprecipitated under the above conditions twice more. The Ni^{57} with its 0.1 to 4 milligrams of nickel carrier was finally converted to NiS

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[1] S. C. Curran, A. L. Cockroft, and J. Angus, Phil. Mag. 40, 53, 522, 631, 929 (1949)

[2] W. Bernstein, H. G. Brewer, Jr., and W. Rubinson, Nucleonics 6, #2, 39 (1950)

[3] G. T. Seaborg and I. Perlman, Rev. Mod. Phys. 20, 585 (1948)

[4] F. Maienschein and J. L. Meem, Jr., Phys. Rev. 76, 899 (1949)

and mounted on thin aluminum foil.

Absorption of the radiations with detection by end-window and windowless counters demonstrated the presence of a component, probably Auger electrons, completely removed by aluminum 0.2 mg/cm^2 thick. The aluminum end-point of the positron activity was at 263 mg/cm^2, equivalent to 0.78 Mev as determined by Feather analysis with Na^{22} (0.552 Mev)[5] as the standard. A small X-ray contribution to the Geiger counting rate was corrected for with the aid of the result of a beryllium absorption experiment. Measurements with a mica-window counter in a magnetic field gave no evidence for the presence of negative electrons. Examination of the radiations from a source approximately 0.5 mg/cm^2 thick in a lens type beta ray spectrometer showed the positron energy to be 0.845 ± 0.010 Mev. A line of electrons, very weak in intensity (less than one percent of the positron activity) was observed at 114 kev. A Kurie plot shows that the positron spectrum is simple and has an allowed shape.

The source was not sufficiently active to allow detection in the spectrometer of the gamma rays accompanying the decay of Ni^{57} by means of photo-electrons ejected from a radiator. The energy of some of the Ni^{57} gamma rays was determined to be between 1.7 and 2.2 Mev by the facts that they produce photoneutrons in beryllium but not in deuterium. The half-thickness for the absorption of these gamma rays in lead is 15.7 g/cm^2, which corresponds to an energy of 2.0 Mev. Measurements were made also with a variably biased scintillation counter; by comparison of the bias curves for the gamma rays of Ni^{57}, Pr^{144} (2.19 Mev)[6], Sb^{124} (1.7 Mev)[3] and Co^{60} (1.17 and 1.33 Mev)[3], the energy of the Ni^{57} gamma rays was determined to be 1.9 ± 0.1 Mev. in agree-

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[5] P. Macklin, L. Lidofsky, and C. S. Wu, Bull. Am. Phys. Soc. <u>25</u>, #1, 10 (1950)

[6] D. E. Alburger, et al, Brookhaven National Laboratory Report 39 (AS-3), 17 (1949)

ment with an earlier measurement[4].

The beta and gamma activities were followed separately for 10 to 15 days. They decayed with half-lives of 36.2 ± 1 and 36.6 ± 1 hours respectively. In the spectrometer it was determined that the 114 kev line decays together with the positron activity.

It was found also that cobalt K X-rays are emitted in the decay of Ni^{57}. The counter for these measurements was made from a four inch diameter brass tube, 12 inches long, in which was placed a coaxial tungsten wire 0.004 inch in diameter. Gas composition was 90 percent argon and 10 percent methane; the total pressure was one atmosphere. Radiations entered the sensitive space through a beryllium window 20 mg/cm^2 in thickness. This instrument was operated as a proportional counter, and it demonstrated a voltage plateau in total number of pulses above noise level. The sliding channel analyzer used to measure the pulse spectrum from the counter has been described[2].

If monochromatic X-rays are allowed to enter the counter and if the number of pulses observed in each pulse height interval is plotted versus the pulse height, two maxima are observed in the resultant curve: one whose position corresponds in pulse height to the energy of the incident quanta and the other, much less pronounced, to an energy lower by the K ionization energy of the principal counter gas. The observed peak position for Ni^{57} was compared with the peak positions for K radiations of manganese, copper, and cobalt produced in the decays of Fe^{55}, Zn^{65}, and Ni^{59} respectively. It is clear (Table I) that cobalt X-rays are emitted in the decay of Ni^{57}.

For one sample the decay of the X-ray peak was followed over a period of 12 days. With appropriate corrections for rather small drifts of channel width and peak position, and after subtraction of a long-lived contribution amounting to approximately one percent of the original peak activity, the X-ray half-life was found to be 38 ± 2 hours.

Table I

Identification of X-rays from Ni^{57} Decay

Date	Source of Radiations	Peak Positions (arbitrary units)
1-16-50	Fe^{55}	44.5
	Ni^{57}	51.8
	Zn^{65}	58.0
1-17-50	Fe^{55}	44.0
	Ni^{59}	50.6

The absolute electron capture rate for a sample was determined in the following way. The area under the pulse height distribution curve was measured. A background originating probably from Compton electrons in the counter was subtracted. The net area remaining is equal to the K X-ray counting rate multiplied by the size of the pulse height acceptance interval or channel width. The latter quantity was obtained by two independent methods which gave the same result. One was essentially to put into the analyzer pulses of uniform height from a generator and to see over what range in channel position the pulses were accepted. The other method was to set the area under a curve for Fe^{55} (no gamma rays) equal to the total count observed, i.e., all pulses above noise, multiplied by x, the channel width. For the Ni^{57} sample the counting rate thus calculated was converted to an absolute capture rate by corrections for geometrical efficiency, absorption in air and in the beryllium window, fluorescence yield[7], and counter efficiency. The last quantity, about 0.90 for the counter used, is simply the fraction of the X-rays absorbed by the counter gas in the average path.

For the same Ni^{57} sample for which the electron capture rate was

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[7] R. M. Steffen, O. Huber, and F. Humbel, Helv. Phys. Acta 22, 167 (1949)

measured, and absolute beta disintegration rate was determined by correction[8] of the measured Geiger activity for solid angle, absorption in air and counter window, back scattering, coincidence loss, and X and gamma activities. The ratio of positron decay rate to the simultaneous K-capture rate is 1.0 ± 0.1.

Coincidences were observed from Ni^{57} between counts in a mica-window G-M tube and in a scintillation counter biased so as not to register annihilation radiation. These coincidences, corrected for chance events confirm the observation of Maienschein and Meem[4] that a hard gamma ray follows positron emission. This is presumably the 1.9-Mev ray previously mentioned. The coincidence rate per hard gamma in Ni^{57}, corrected for a few $X-\gamma_h$ events, was compared in a fixed geometry with the same quantity for Na^{22}, which decays by emission of a positron followed by one 1.3-Mev gamma quantum[9].

The ratio

$$\left(\frac{\beta - \gamma_h}{\gamma_h}\ \mathrm{Na}^{22}\right) \Bigg/ \left(\frac{\beta - \gamma_h}{\gamma_h}\ \mathrm{Ni}^{57}\right)$$

is 1.9. Here the symbol γ_h means the hard gamma rays, and $\beta-\gamma_h$ stands for positron-hard gamma coincidences. The result indicates that in the decay of Ni^{57} there are hard gammas emitted other than those associated with the positrons; the result is consistent with the emission of one hard gamma following electron capture also.

Coincidences were measured between a scintillation counter biased as above and one whose bias was set close to noise level. The ratio was found to

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[8] B. P. Burtt, Nucleonics 5, #2, 28 (1949)

[9] W. M. Good, D. Peaslee, and M. Deutsch, Phys. Rev. 69, 313 (1946)

$$\left(\frac{\gamma - \gamma_h}{\gamma_h} Na^{22}\right) \Big/ \left(\frac{\gamma - \gamma_h}{\gamma_h} Ni^{57}\right)$$

be 1.2; it should have been approximately 2.0 if the only gammas other than the hard ones were from the annihilation of the positrons. This observation of gammas in cascade is in agreement with the work previously cited[4]. The number of gamma rays in cascade was not determined because the efficiency of the scintillation counter for the softer gammas was not known. An increase in the bias of the lower-biased counter made it possible to cut off the softer nuclear gamma, although sensitivity to annihilation radiation remained. Under this condition the ratio

$$\left(\frac{\gamma - \gamma_h}{\gamma_h} Na^{22}\right) \Big/ \left(\frac{\gamma - \gamma_h}{\gamma_h} Ni^{57}\right)$$

was found to be 2.0 in good agreement with the $\frac{\beta-\gamma_h}{\gamma_h}$ experiment. It is thus established that the softer gamma ray has an energy less than 0.5 Mev as Maienschein and Meem suggested. This soft gamma may or may not be the 120 kev gamma whose conversion electrons were observed in the beta-ray spectrometer.

In an attempt to ascertain whether gamma rays are in cascade in the positron branch of the Ni^{57} decay, coincidences were measured for Ni^{57} and for Na^{22} between a G-M tube and a scintillation counter biased just above noise level. The counters were shielded from each other by a one-inch thickness of lead. Measurements were made both with and without a beryllium absorber for the positrons in front of the mica window of the G-M tube, so that it was possible to correct for gamma and X-ray counts in the G-M tube and for X-γ coincidences. The ratio

$$\left(\frac{\beta - \gamma}{\beta} Ni^{57}\right) \Big/ \left(\frac{\beta - \gamma}{\beta} Na^{22}\right)$$

was calculated to be 1.2. The $\frac{\gamma - \gamma_h}{\gamma_h}$ result shows that the annihilation quanta and softer gammas of Ni^{57} contribute about equally to the $\gamma - \gamma_h$ coincidence rate and therefore equally to the single rate in the low biased counter. It would thus appear from the value 1.2 for the ratio

$$\left(\frac{\beta - \gamma}{\beta} Ni^{57} \right) \Big/ \left(\frac{\beta - \gamma}{\beta} Na^{22} \right)$$

that relatively few of the soft gammas of Ni^{57} are in the positron branch of the decay. In fact, because the 1.9 Mev Ni^{57} quanta are counted more efficiently than the 1.3 Mev Na^{22} quanta, the ratio should be greater than 1.0 even if no gammas are in cascade in the positron branch of the Ni^{57} decay. Thus it may be concluded that very few, if any, of the positrons are followed by a gamma ray cascade; this result agrees with the simple shape of the Kurie plot for the positrons.

The results of this investigation are consistent with the decay scheme diagrammed in Fig. 1. The number of gamma rays in cascade in the capture branch is not known. But if additional gamma reys esist in this branch their energy must be < 0.5 Mev.

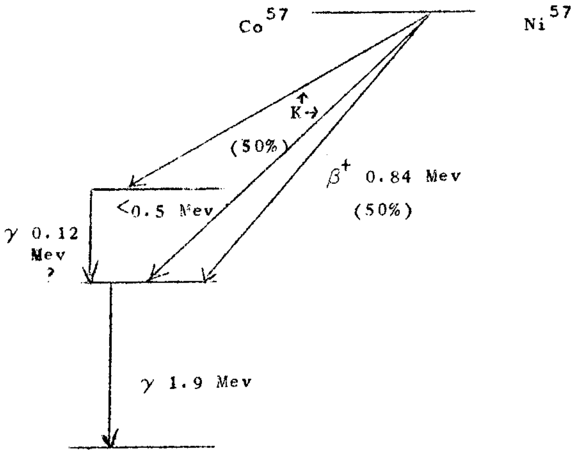

Fig. 1. Decay scheme of Ni^{57}

We are indebted to Dr. D. Cowie and Dr. P. Abelson who were kind enough to carry out the cyclotron irradiations.

(G. Friedlander and M. L. Perlman)
(Beta-ray spectrometer measurements - D. Alburger)
(Hard gamma ray energy determination by scintillation counter and photoneutron measurements - E. der Mateosian)
(Coincidence measurements - G. Friedlander and M. L. Perlman in collaboration with A. W. Sunyar)

INSTRUMENTATION & HEALTH PHYSICS DEPARTMENT

ELECTRONICS & INSTRUMENTATION DIVISION

Stable High Voltage Supply:

For some time we have felt a need for high voltage supplies with exceptional long term stability to be used in energy measurements with proportional and scintillation counters. In these applications it is important to keep the overall gain of the system constant for the duration of the experiment, which may be several hours, and since the gain of a proportional counter or photomultiplier is an exponential function of the voltage, extremely low drift is essential. We have developed a supply with an output variable from 1 to 5 Kv which will not vary more than ± .01% from the assigned value for several hours. The reference voltage is obtained from three type 5651 gas tubes operated at constant voltage from a separate supply. Unusually high gain is employed in the regulator amplifier and pains are taken in choice of components and mechanical layout to reduce temperature effects. The output voltage is set by a precision decade potentiometer direct reading in one volt steps, protected with heavy thermal insulation. Provision is made for calibration relative to a standard cell. Three of these supplies have been delivered and the same principles have been employed in a lower voltage supply for an ionization chamber and amplifier now under test.

Precision Timer:

Work has started on precision timing equipment for the time-of-flight mass spectrometer proposed by Dr. Goudsmit (Physics). The ion source of the spectrometer will be pulsed and it is desired to measure the time of flight of the ions to 1 part in 10^5. The timing circuits will be similar in principle to those used in Loran. The standard will be a 1 mc or 10 mc crystal with frequency dividers reducing this to the repetition rate of 100 cycles and phase shifting to interpolate down to .01 microseconds. We have developed a ring of 10 scaler circuit for the frequency division which works up to 1.5 megacycles. Each ring consists of 10 duo-triode flip-flops. Each flip-flop is connected to the next and to the input signal so that if one stage is in state A and the others are in state B, the non-symmetrical state A will be passed from stage to stage by each successive input pulse in the conventional manner. If more or less than one stage gets into the A state, the ring will divide by the wrong factor.. Stability is assured in this circuit by biasing of the cathodes. The cathodes are self biased; all the cathodes on one side of the flip-flops going to one common cathode resistor and the opposite side to another. The side which should have 9 "on" tubes has one value of cathode resistor and the side which should have 1 "on" tube has another 9 times as big. The only stable state is one in which there is one stage in state A. All trigger signals are connected to the appropriate plates through hard diodes. The fast stage uses 6AL5 and 5687 tubes; the others 6AL5 and 12AU7s.

Particle Range Sorter:

Dr. H. Motz (Physics) has built a multiple wire proportional counter for energy discrimination of heavy particles. A collimated beam enters one side of the counter. The path length in the gas of the counter will be determined by locating the wire at which the tract ends. Electronics built the circuits which consist of 11 amplifiers, one for each wire. There are 2 feedback loops with a gain of about 10^4 and rise time of 0.2 microseconds. Each amplifier is connected to a discriminator. Anti-coincidences are taken between successive channels and coincidence with the first detector. A scale of 4 and register completes each channel.

GM Tubes for Liquid Flow:

The Reactor Division is making a study of the dynamic behaviour of an ion exchange column. Special GM tubes were constructed, in which the liquid flows through a fine glass spiral inside the counter volume. Two standard ratemeters were revised to work with these counters in this experiment.

Scintillation Counters for Civil Defense Monitoring:

We have done some investigation of the possibilities of scintillation counters for civil defense monitoring purposes. Several different crystals, including calcium tungstate and sodium iodide, appear satisfactory, and any of a large group of 931A photomultipliers can be used with an adjustment of the voltage. For indication we could use a cold cathode trigger tube, WL759. The photomultiplier current charges a condenser which discharges through the trigger tube after reaching a definite potential. The occurrence of the discharges can be indicated visually by a flashing light or meter deflection, or aurally with headphones. This scheme should be free from saturation effects at any radiation levels likely to be encountered, and would have long life. Unfortunately the WL759 is not in commercial production so we cannot form an accurate estimate of the cost of a complete unit.

Atmospheric Potential Gradient Recorder:

An instrument for recording the atmospheric potential gradient has been designed for the Meteorology Group, who plan to try using it to determine when the plume from the reactor stack is overhead. In this device a carefully insulated radioactive probe extending several feet horizontally from the case is connected to one terminal of a vibrating condenser. The alternating e.m.f. resulting from any potential across the condenser is amplified, synchronously rectified, and applied to the other terminal of the condenser, and to a shield around the probe. Thus the potential of the shield is driven to closely approximate that assumed by the probe, with the result that the atmospheric gradient is essentially undisturbed. The model now under test will follow potential changes (with respect to the case) somewhat over plus or minus 200 volts, corresponding to gradients of plus or minus 1000 volts/meter, and records directly on a 1 m.a. Esterline-Angus meter.

Publications:

Papers published in the last three months by members of the Electronics Division are:

"Desirable Improvements in Electronics Instruments"
J. B. H. Kuper, Nucleonics Vol. 6, No. 2., p. 26, Feb. 1950

"A Proportional Counter X-ray Spectrometer"
Bernstein, Brewer and Rubinson (Chem.), Nucleonics, Vol. 6, No. 2,p.39 Feb. 1950.

"Gas Phase Counting of Low Energy Beta Emitters"
W. Bernstein, R. Ballentine (Biol.), R.S.I. 21, 158, Feb. 1950

"Measuring a Varying Frequency"
R. L. Chase, Electronics, Vol. 23, No. 3, P. 110, March 1950

Also, the following papers are in press:

"A Monitor for Low Intensity Gamma Rays"
J. B. H. Kuper and R. L. Chase, R.S.I.

"A Monitor for Airborne Radioactive Dust"
J. B. H. Kuper, E. H. Foster and W. Bernstein, Nucleonics.

HEALTH PHYSICS DIVISION

A one week training course in health physics for representatives of casualty insurance companies was given in February. There were 92 in attendance from 42 companies. Comment received from the companies has been very favorable.

Another major project of this period was preparation for the training course for instructors in Radiological Monitoring for Civilian Defense. A five week curriculum of classroom, laboratory and field activities has been planned in full detail. A total of 25 speakers, including 12 from outside the laboratory, has been obtained.

Normal health physics services have been maintained. The chief services activities have been concerned with pile testing, breaking-in of the Van de Graaff generator, and operation of the Chemistry hot laboratory and the Waste Processing Laboratory. Plans for sampling and monitoring equipment at the liquid waste disposal plant have been completed and the construction contracts let. Collection of background data at the area monitoring stations has been continued with a record of 84% apparatus efficiency.

Organization of a Disaster Team to provide radiological monitoring in the event of an offsite accident involving radioactivity is well under way. Monitoring equipment has been obtained from the AEC stockpile and is being put in working order, and arrangements have been made to use the Laboratory Fire Department Rescue truck and two jeeps for transport. Training of the team members can commence as soon as funds are made available for tires, radio equipment and miscellaneous supplies for the truck and jeeps. Pending the training of a state civil defense monitoring service, it is expected that the Laboratory team will be ready to serve if and when called upon by the appropriate authority.

MEDICAL DEPARTMENT

During the past three months organizational development has proceeded in the medical department.

Division of Biochemistry

Laboratory work in this division has comprised largely the development of methods suitable for patient study. Several more methods are yet to be learned before work planned can get started with any momentum. The counting laboratory and biophysics section of the division is being organized and should be in operation within another month.

Division of Bacteriology

This division has several problems of considerable interest. One of these investigations, on the effect of gamma radiation on the various immune processes and mechanisms of infection, has required that first the lethal dosage of radiation from cobalt-60 be established for the infection-free colony of mice used by the division. It is known that this differs from the number of r required by ordinary x-ray and it was deemed advisable to determine the lethal dose here rather than use values available in the literature, because of strain differences. All radiations referred to here were from a 2 curie source of cobalt-60. The results of these are shown in the accompanying table.

Autopsies on mice sacrificed just before death from 1333 r dosage showed no subcutaneous or mesenteric hemorrhage. The intestines were distended, and no formed stools were present. The mice had some diarrhea. The spleens had all but vanished, and cultures on the hearts' blood, spleens, and livers produced no bacterial growth, which indicated that the mice were not dying of infection. Mice receiving 2000 r showed a similar picture, except the intestines were discolored, but did not appear to be bloody. The fact that the cultures were negative and that penicillin and aureomycin do not seem to prolong the life of the mice seems to indicate that these "infection-free" mice do not have infection in the acute radiation sickness, which may be the reason for their apparent resistance to gamma radiation as compared to the usual findings. However, more observations need to be done before this is definitely established.

The numbers of mice in the accompanying table are far too small for any statistical evaluation, but due to the very limited capacity of our present cobalt source, no larger group could be used. The L.D. 50 dose will be determined for the different age groups when a larger source is obtained. However, it is obvious that there is a sharp difference between the 21 and 31 day old mice. Mice 10 months old are now being radiated to compare them to the younger mice.

Division of Pathology

This division is extending studies on the effects of injury on protein metabolism.

The Hospital

During this quarter the hospital completed its first year of operation. All that could be hoped for from this year of experience has been gained. The unit is a smoothly functioning one ready for whatever problem any patient may present. The credit for this development must be given largely to the nursing and adjuvant staffs of the hospital.

In the hospital the steady growth in number of patient days has continued. Satisfactory base lines have been obtained on most of the research patients so that in most instances more active investigation can begin in the near future. The patients remain approximately one-third carcinoma of the thyroid and two-thirds non-cancer.

TABLE

NUMBER & SEX OF MICE	AGE	GAMMA RADIATION DOSAGE	ANTIBIOTICS ADMINISTERED IMMEDIATELY AFTER RADIATION & DAILY FOR 3 DAYS*	RESULTS	BACTERIOLOGICAL EXAMINATION OF HEARTS' BLOOD, OF SPLEEN & LIVER
12 ♀	4 weeks	546 r	none	ill 3rd and 4th day	not done
12 ♀	8 weeks	640 r	none	mild illness 3rd and 4th day	not done
15 ♂	21 days	1050 r	none	** D_4 x 3 D_5 D_8 D_{11} D_{13} D_{14} x 2 D_{16} x 2 D_{18} D_{19} D_{42}	not done
12 ♂	31 days	1050 r	none	D_9	not done
12 ♂	12 weeks	1080 r	none	very mild illness 4th day testicular atrophy 21 days	not done
12 ♀	4 weeks	1333 r	none	D_3 D_4 x 2 D_5 D_7 D_9 D_{10} x 2 D_{11} x 2 D_{13} D_{14}	no growth
12 ♀	4 weeks	1333 r	P in H_2O 500 u S in H_2O 500 u P in oil 30,000 Aureo. 1 mg. per mouse	D_3 x 3 D_6 x 4 D_8 D_9 x 2 D_{10} D_{11}	no growth
12 ♀	4 weeks	2000 r	none	D_3 x 12	no growth

*Streptomycin and Penicillin in H_2O given only immediately after radiation. Penicillin in oil and Aureomycin given daily for first 3 days.

**D6 = 1 died 6th day following radiation.
D_6 x 6 = 6 died 6th day following radiation.

NUCLEAR DYNAMICS

Isomerism in Ti^{51} and Nuclear Shell Theory:

The nuclear shell theory of M. G. Mayer makes predictions concerning the distribution of isomeric states throughout the periodic table. Two reported cases of isomerism, Ca^{49} and Ti^{51} were in disagreement with her predictions.

The lifetimes of the isomeric states of Ca^{49} as recorded in the literature were 2.5 hr. and 30 min. These activities were studied at the Argonne National Laboratory in 1948 and both found to be falsely attributed to Ca^{49}. This nuclide has only an 8.5m activity. It was also found that a 72d activity attributed to Ti^{51} was excited with less than 1/10 the cross-section attributed to it at the time. Recently a 72d Ti^{51} source was obtained from Oak Ridge and subjected to a very careful chemical treatment. Proportional counter and photo neutron detector studies indicated that large percentages of the activity of the sample were Sb^{124}(60d) and Hf^{181} (46d). By repeated purifications, Dr. Miskel reduced the specific activity of long lived activities in Ti to 1 part in 250,000, making the existence of an isomeric state in Ti^{51} highly unlikely. (M. Goldhaber, E. der Mateosian, J. Miskel)

"Memory" in Angular Correlation of Gamma Rays From Decay of Pb^{204}:

Following K capture in Bi^{204} an excited state of Pb^{204} is formed. This state decays with a 68 minute half life by the emission of a 905 kev partially converted gamma ray to a state of 374 kev excitation energy. This state subsequently decays to the ground state of Pb^{204} with a lifetime of about 3×10^{-7} seconds by the emission of a partially converted gamma ray. Probable multipole orders for these radiations were previously reported on the basis of lifetimes, conversion coefficients and K/L ratios. In an effort to check spin assignments, experiments have now been performed to measure the directional correlation of the two gamma rays using delayed coincidence techniques with scintillation counters as detectors for the gamma rays. A source of lead was bombarded with high energy deuterons in the M.I.T. cyclotron. The bismuth activity was chemically separated from the bismuth fraction. Two sets of coincidence measurements were then made with gamma ray counters 180° and 90° apart in angle. One counter was delayed about 5×10^{-7} seconds with respect to the other. A directional correlation was observed at this delay. This is the first direct evidence that a nuclear excited state retains memory of its spin direction for a time as long as 0.5 microseconds. Our measured ratio for the number of delayed coincidences at 180° to the number at 90° is 1.2 ± .05.

(M. Goldhaber, A. Sunyar, G. Friedlander)

Hard Gamma Rays:

The following is a tabulation of all new hard gamma rays which have been found to give photo neutrons in Be or D. The γ-ray energies listed in column 2 for Bi^{206} and Ce^{144} were measured by D. E. Alburger with a lens spectrometer. Ni^{57} has been studied by Drs. G. Friedlander and M. Perlman (see report in Chemistry section) who detected the hard γ-ray by lead absorption. In this case, photo neutrons were observed with Be but not with D_2O. The energy of the gamma ray was measured by comparison with the 1.7 mev γ-ray of Sb^{124} and the 2.185 mev γ-ray of Pr^{144} using a scintillation counter γ-ray spectrometer.

PHOTO NEUTRON INTENSITIES

Source	γ Energy (Mev)	Relative Intensities of Photo Neutrons From Be	From D_2O
Ra	several from 1.69 to 2.42	1000	50
Ag^{110} (225d)	$1.67 < E\gamma < 2.23$	0.5	-
Bi^{206} (6.3d)	1.72	330	-
Ce^{144} (275d)→Pr^{144} (17m)	2.185	650	1
Ru^{106} (1y)→Rh^{106} (30s)	$2.23 < E\gamma$	150	15
Ni^{57}	1.9 ± 0.2		-

(M. Goldhaber, E. der Mateosian)

Beta Ray Spectrum of K^{40}:

Continuing the studies of highly forbidden beta spectra, a measurement of the beta-ray spectrum of K^{40} has been made using a lens spectrometer at 17 percent resolution. The source was prepared under the direction of C. P. Keim by electromagnetic enrichment at the Oak Ridge Y-12 plant. The K^{40} content is 7.13 percent. Enriched KCl was deposited by precipitation over a 4 cm^2 area on 0.5 mg/cm^2 Nylon backing. The average thickness of the source was 2.4 mg/cm^2. A maximum net counting rate of 16 per minute was observed above a background of 18 per minute. Calculated correction for resolution was made by graphical integration. The Kurie plot, corrected by the 3rd forbidden axial vector or tensor interaction factor for spin change 4, is linear from about 500 kev to the end-point at 1.36 ± 0.03 Mev. Other forbidden transition factors have been applied to the data. α (first forbidden correction factor) appears to give a non-linear plot but a D_2 correction (second forbidden) is linear above 500 kev and cannot be excluded with present data. Deviations below 500 kev have been shown to be due to effects of source thickness by comparison studies using the allowed beta emitter P^{32} mixed in normal KCl.

(D. E. Alburger)

Neutron - Deuteron Collisions

Angular distribution of deuterons recoiling from 4.5 and from 5.5 Mev neutrons was reported at the New York meeting of the A.P.S. (February 1).

Since then the measurements have been extended to larger angles, with improved statistics, and the correctness of the n - D measurements has been assured by additional measurements of neutron - proton recoils in the same apparatus, this latter work agreeing with the accepted n - p symmetrical distribution.

The results are definite enough to show that the only extant theoretical computation of n - D scattering at these energies is not in agreement with the measurements. Some modification of the theoretical calculation is necessary, and contributions by d - waves to phase shifts are suggested.

(E. Wantuch, E. O. Salant)

Photo - Disintegration of Rhodium

The preliminary work, published in the January 15 issue of the Physical Review, has been extended by measurements of more photoprotons at the same angles and by measurements at different angles. Except that the accuracy of the work is improved, the errors having been reduced more than a third, the general conclusions of the publication, i.e., the value of the threshhold, the excitation curve, the cross section, and the predominance of right - angle ejection for the photoprotons of higher energy, remain as published.

(N. W. Curtis, J. Hornbostel, D. W. Lee, E. O. Salant)

Fast Neutron Cross Sections and Nuclear Shells

Measurements (begun at Argonne National Laboratory) of capture cross sections for fast neutrons are being continued with special reference to nuclei with neutron number near the magic numbers 50, 82, and 126. A low cross section is interpreted as a low neutron binding energy and the discontinuities in cross section at the magic numbers of a factor of about 100, necessitate a shift in binding energy of about 4 mev. Isotopes now under study are long lived, have been bombarded at Argonne, and are being counted at Brookhaven. Re^{185} (110 neutrons) and Re^{187} (112 neutrons) have been found to have cross sections about 150 mb and hence normal neutron binding energies; but Ru^{102} (58 neutrons) has only about 6 mb, hence a low binding energy. Ru^{102}, and Mo^{100} (58 neutrons) already reported by us as low, provide evidence for a closed sub-shell at 58 neutrons.

(D. J. Hughes and R. Garth.)

Particle Range Sorter

A ten channel counting system for sorting protons, deuterons, and alpha particles according to their range is being constructed for the study of reactions produced with the Van de Graaff Generator. The instrument consists of twelve wire to plane proportional counters each followed by a linear amplifier. Coincidence and anti-coincidence circuits are employed between these twelve channels such that only the particles entering through the first counter and ending in a particular counter are recorded in a given channel. In this manner particles whose ranges fall in intervals R_i and $R_i + \Delta R$ for ten adjacent values of R_i are simultaneously observed. The counting interval ΔR for each counter is determined by the counter dimensions and filling. The design results in stable operation with high discrimination against background and allows the rapid accumulation of data.

The electronic circuits have been completed by the Electronics Division and are being calibrated. A model consisting of four counters is being tested to aid in the design of the twelve section counter. A foil changer system and air absorption cell will be installed between the counter and the source of radiation to permit detailed scanning of spectra over a wide range. (H. Motz)

Time-of-Flight Velocity Selector to Operate on Neutrons Coming from the Pile With Energies Between 1 EV and 100,000 EV

The neutron shutter (chopper) for this machine has been under consideration. Two designs have evolved; viz.:

(I) Vertical Spinning Rotor; (II) Horizontal Spinning Rotor

In both cases, there would be an arrangement of stationary collimating slits which define four parallel horizontal neutron beams. Either rotor would contain one or more sets of slits which would periodically (i.e. every 300 μ secs or so) become aligned with the stationary slits allowing the simultaneous passage of bursts of neutrons, one associated with each neutron beam. However, in the first case the rotor would be about 24" in diameter and would be spun about a vertical axis normal to the direction of the neutron beams; while the other rotor would be not more than about 8" in diameter and would be spun about a horizontal axis parallel to the direction of the neutron beams.

It has become obvious that pains will have to be taken to properly shape and place the rotor slits in both designs. Apparently the neutron counting rates resulting from use of either design of rotor are about the same with perhaps a slight advantage in favor of the vertically spun rotor. It should be pointed out that the operating speed of the latter will not exceed 20,000 RPM; on the other hand, if the horizontal rotor were to be used, it will be desirable to attain 50,000 RPM. The goal for this instrument is to obtain a resolution of the order of 0.1 μ sec/m and to be able (perhaps at slightly poorer resolution) to study samples of cross-sectional area the order of 0.01 square inches.

The final decision as to which design will be best now seems to depend upon experimental tests.

(F. G. P. Seidl, D. J. Hughes)

Time-of-Flight Neutron Spectrometer Using Pulsed Cyclotron

The electronic timing and routing circuits being built under a contract with Columbia University, are about 80% completed.

(C. P. Baker)

HIGH ENERGY PARTICLE PHYSICS

Altitude and Latitude Dependence of Bursts in a Lead Shielded Ion Chamber

The instrument used in this experiment consists of a cylindrical ionization chamber covered by a half cylindrical lead shell 15 cm thick and of 6 trays of G-M tubes, of which 5 are placed around the lead shield and one directly under the chamber. Various kinds of coincidences between bursts of the ionization chamber and pulses of the counter trays are recorded. These events are interpreted as due to nuclear interactions of cosmic rays. The instrument was installed aboard a B-29 and flown at the pressure altitudes of 273, 300 and 383 g cm^{-2} at 55°N geomagnetic latitude, and at the pressure altitudes of 300 and 383 g cm^{-2} at 20°N geomagnetic latitude. The experiment provided information on the altitude, latitude and angular dependence of the radiation responsible for nuclear interactions. If one considers in particular penetrating ionizing particles capable of producing high energy nuclear events after traversing 15 cm of lead, one finds that the altitude dependence corresponds to an absorption thickness of about 124 g cm^{-2} both at 55° and 20° and that the intensity ratio between the two latitudes is about 1.14.

(Allen J. McMahon - Bruno Rossi, M.I.T.)

Energetic Stars at Different Latitudes

Eastman NTB 3 emulsions were flown to an atmospheric pressure of 15 gms/cm^2 at 57° and 31° north geomagnetic latitudes, cut-offs 2.8 and 8 Bev respectively. About 1000 stars were examined in plates verified as sensitive to minimum ionization tracks. Latitude effect for star production by incident singly charged, relativistic particles is about the same as reported for particle flux by Winckler et al. (private communication), implying approximately constant cross section, over relevant energy ranges, for proton disintegrations. No significant differences between the two latitudes are found for distribution in either numbers of heavy prongs or multiplicities of outgoing minimum tracks. Considering only stars of 3 or more heavy prongs, data at each latitude show:- (a) about 1/4 of the stars are caused by relativistic protons, (b) 2/3 of the proton-induced stars have outgoing minimum tracks, presumably mostly relativistic mesons, with an average multiplicity of 3, (c) 2/3 of these stars have 6 or more and 1/2 have 9 or more heavy prongs, (d) 1/2 of all stars contain only heavy prongs, of which only 1/4 have 6 or more prongs. This distribution, contrasted with (c), suggests only moderately energetic neutrons as a cause.

Features of the high energy disintegrations revealed by the analysis include the following: (1) the light nuclei in the emulsion disintegrate principally into 2,3 and 4, and occasionally into 5 heavy prongs, (2) the mean multiplicity of emitted particles of relativistic energy (mostly mesons) is the same at the two latitudes, showing that the probability of meson creation by particles in the energy range 2 - 20 BeV is essentially independent of the energy of the particles, (3) the multiplicity of emitted energetic particles increases with the size of the nucleus struck, this increase being closely the same at the two latitudes. These data lead to the conclusion that creation of many mesons in a collision of a proton or neutron with a heavy nucleus arises from many collisions as a particle passes through the nucleus, rather than from "multiple" production of the mesons in a single nucleon-nucleon collision. If the conclusions of Fowler of Bristol are accepted, then our results mean that only one meson is created in a nucleon-nucleon collision, but it should be noted that it is difficult to reconcile Fowler's conclusions with certain other cosmic ray data. It is, indeed, feasible, at the present time, to interpret Fowler's data and our observations, that 2 mesons are created in a single nucleon-nucleon encounter; whichever way the current studies may decide this point, the creation of many mesons by many collisions of a particle passing through a nucleus, the so-called "plural" production, seems established. (C. B. Fisk, J. Hornbostel, E. O. Salant and J. E. Smith)

High Pressure Cloud Chamber

After the cloud chamber and controls had been completed, difficulties with fogging of the cloud chamber were encountered until the cause of the trouble was traced to electric conductivity across the wall of the cloud chamber. After reducing the current by improved insulation to approximately 1 microampere at 3,000 Volt of sweeping field, the chamber started to work satisfactorily. First, helium up to 3,000 psi was used. Satisfactory pictures were obtained with a cycling time of 20 minutes. However, after the time for recompression of the cloud chamber was reduced from 20 seconds to 6 seconds, good pictures could be taken every 5 minutes. At the present time, pictures are being taken with argon at 4,200 psi, where again the cycling time has been reduced from 30 minutes to 10 or 15 minutes by reducing the time for recompression from 18 seconds to 6 seconds. Some difficulty is still encountered with fogging of the front plate and turbulence. Thermostating of different sections of the chamber is therefore being installed. So far, one stopped meson and one three-pronged star have been obtained out of 35 pictures taken with argon. A magnetic field of 8,000 gauss can now be applied by means of lead storage batteries until the Diesel generators are ready for operation.

(R. P. Shutt, E. C. Fowler, T. H. Johnson)

A High Speed Expansion Valve for Cloud Chamber Control

Whenever accurate measurements are to be performed on a cloud chamber photograph, it is important to keep the width of the tracks to be measured down to less than 1 mm. Considering ion diffusion, one finds that in air or argon the time delay between passage of a ray through a cloud chamber and completion of an expansion must be 10^{-2} sec or less. Some difficulties in designing a sufficiently fast acting expansion control valve may be encountered when a large cloud chamber is to be controlled by Geiger counters. During the short time indicated the magnetic field in the holding electromagnet usually employed must be decreased to a point where the valve starts to open, a movable valve disc must fly open, and air from the back compartment of the cloud chamber must rush out through the valve opening. For a large cloud chamber a large valve opening is required, often making the valve heavy and slow-moving and resulting in a long time constant of the magnet. Although these difficulties have been overcome in many different ways by other workers, a review of all the factors involved made it desirable to construct an expansion valve combining a number of principles not all of which have been used by others. By balancing pressures the movable valve parts can be made very light despite the large diameter of the valve opening, and the holding magnet can be quite small. Details of the design are described in an article for the Review of Scientific Instruments. Approximate calculations were made concerning all the important design features. Because of their general applicability some of these calculations are also reproduced in the article. An important result is that even a small electromagnet must be laminated to insure sufficiently fast decay of its magnetic field. It is of interest also to note that most of the remaining time delay is caused by the aerodynamic resistance at the valve opening. For the present design employing a valve opening of 2.8 cm radius expansions are completed 8×10^{-3} sec after passage of the initiating ray. This agrees with the calculated delay. Electric power consumption of the electromagnet is only 0.17 Watt so that a small vacuum tube is sufficient for operation. It should be pointed out that the principle of pressure balance utilized in this valve would enable one to construct a much larger valve operating as fast as the valve described here with only a slight increase in power consumption.

(R. P. Shutt and W. L. Wittemore.)

Construction of a Large Permanent Magnet for Cloud Chamber Work

For the investigation of cosmic rays, one can arrange a magnetic field between two cloud chambers. For the same flux of radiation to be observed, the air gap of the magnet can be made much smaller in this arrangement than when a cloud chamber is mounted between the pole pieces of a magnet. For small gap widths a permanent magnet is capable of producing a strong magnetic field. Since angular deflections of rays passing through the cloud chamber and the magnetic field are to be measured, the product of the magnetic field intensity H in the gap and the height h of the pole faces must be made a maximum in order to enable one to measure large momenta. In order to assure optimum amount and distribution of the magnetic alloy used to produce the permanent field, model experiments were performed. Procedure and results are reported in an article for the Review of Scientific Instruments. The most important result is that for a given amount of alloy the product Hh becomes largest when the alloy is concentrated as close to the pole faces as possible, while H alone is not sensitive to the distribution of the alloy. The proper amount of taper at the pole faces is also found by the experiment. The magnet constructed has pole faces 11.3" square, a gap 1.25" wide, and produces a maximum field of 9,000 gauss.
(W. L. Wittemore and R. F. Shutt)

NEUTRON OPTICS AND SOLID STATE

Coherent Neutron-Proton Scattering by Liquid Mirror Reflection

An accurate determination of the coherent neutron-proton scattering amplitude, a_H, is important because of the direct connection between a_H and the fundamental nuclear force constants. A value for a_H of $(-3.75 \pm 0.03) \times 10^{13}$ cm was recently obtained by a comparison with carbon, involving critical reflection of neutrons from a liquid hydrocarbon ($C_{12}H_{18}$) mirror. The experiment has now been repeated, using various liquids and a range of neutron wave lengths, in order to search for systematic errors. With $C_{12}H_{18}$, the unbalanced coherent scattering amplitude (a_C is positive and slightly larger than 1.5 a_H) was determined from the measured wave length at a fixed incident angle (about 5 minutes). The present method consists of a determination of the particular C-H ratio for which the amplitude is zero, at which ratio the reflected intensity is zero for all incident angles. The reflected intensities from liquids of different C-H ratios, some of which are mixtures, are measured at different angles, and it is then a simple matter to extrapolate to the C-H ratio corresponding to zero amplitude. The values of a_H obtained with different liquids and neutron wave lengths are in agreement with the $C_{12}H_{18}$ result and make systematic errors unlikely.

(D. J. Hughes - M. T. Burgy and G. R. Ringo, Argonne National Laboratory)

Oriented Overgrowth of Alkali Halides on Metal Crystals

Since the January report several new observations have been made of the crystalization of alkali halides on metal surfaces which indicate that closeness of fit of the two lattice spacings cannot be regarded as a strictly required condition for oriented growth. NaCl and RbCl, whose lattices differ by 15% were both oriented on cleaved Bismuth; NaCl was oriented on the (111) plane of both silver and copper whose lattice spacings differ by 10% and oriented growth occurred on almost every grain of polycrystaline silver regardless of the indices of its surface plane. That the orientation of the overgrowth was determined in the embryonic stages of growth is suggested by the observation that crystals often span the boundary between surfaces of different indices without modification.

(G. W. Johnson)

THEORETICAL PHYSICS

Quantum Field Theory

A basic difficulty in the theory of interaction of elementary particles is the fact that solutions of the quantum mechanical field equations usually appear to be intrinsically divergent. In a paper for publication in the Physical Review, it is attempted to show that the divergence is due to the form of the perturbation calculations generally employed and not to the Field Theory itself. By means of a minor modification in the perturbation calculations, the divergent integrals may be given appropriate finite values. Although there are many apparent ambiguities in Field Theory, these are ambiguities of representation, and not abiguities in physical content. The self-energy and stress of particles are zero. The expectation value of the energy momentum tensor for the true vacuum state is zero. From this point of view the polarization of the vacuum and charge renormalization seem to be factitious.
(H. S. Snyder)

Scattering Induced Curvature of Fast Charged Particles

The scattering of charged particles while passing through matter presents a limitation to the accuracy with which the momentum of the particle can be determined by the method of magnetic bending in a cloud chamber, but it also gives useful information for identifying particles from the character of their tracks in emulsions. The exact solution recently obtained by W. T. Scott and H. S. Snyder for the small angle plural and multiple scattering of fast charged particles is used in a recent paper for the Physical Review to derive several theorems of interest concerning scattering-produced curvatures, and calculations are reported of the probability distribution of such curvatures. Formulas are provided for obtaining the probability of occurrence of curvatures greater than any given amount, for any scattering material and energy, as long as the scattering is not too large. The types of curvature measurement dealt with are: a) three-point observation: b) mean curvature using the tangents to the track at each end: c) difference between the two curvatures obtained in (b). Asymptotic formulae allow the calculations to be extended for unusually rare events.

It is shown, first, that knowledge of the probability of a lateral displacement x after a track length z is sufficient to find the distribution of scattering-produced curvatures c obtained by observing the ends of the track and any one interior point. The appropriate relationship is $x = z^2c/2$.

It is further shown that by introducing a lateral displacement angle $\phi = x/z$ the probability of observing ϕ in length z may be obtained by a "duplication formula" from the distribution in ϕ_1 at any $s_1 < z$ and the distribution in $\phi - \phi_1$ in a track of length $z-z_1$.

From the basic correlated distribution formula, distributions have been obtained for the curvatures of the two circles tangent to either end of the track and passing through the other end. The mean $\bar{c}$ of these two curvatures (a special "four-point" curvature) follows the same distribution law as the directional distribution of the previous paper, with $\eta = z\bar{c}$. The difference of these curvatures, D, follows the same law as the three-point curvature c, above.

Finally, it has been shown that the distribution in x and η, or in c and $\bar{c}$, are nearly the same if we write $\eta = \sqrt{3}(x/z)$ or $\bar{c} = (1/2)\sqrt{3}c$, although the exact solution yields different expressions for the appropriate Fourier transforms.

Calculations are here reported and summarized in graphs of the differential and integral distributions in $\phi = x/z$, in terms of dimensionless units z/λ and ϕ/η_c as in the previous work. Integral distributions in angular displacement η, derived from the previous calculations, are included for completeness.

(W. T. Scott and H. S. Snyder)

Dosage From Homogenously Distributed Emitter

In a theoretical study of biological interest, the radioactive dosage received by a biological organ due to a radioactive emitter uniformly distributed therein has been calculated for a wide range of shapes and sizes of the organ. The results, which will be published in Nucleonics, are presented in the form of expressions, valid under various conditions, for the fraction F of the total energy not absorbed in the organ because of escape through the surface. Both radiations which ionize uniformly along their paths and those whose ionization varies universely as their energy have been considered.

(P. I. Richards, B. A. Rubin)

In addition to the following unclassified work of the Department of Reactor Science and Engineering, a classified Progress Report for this quarter (BNL-52) is being issued. This reports progress in Reactor Chemistry, Metallurgy, and on the subcritical experiments on the BNL Reactor.

Studies of the Systematics of Alpha-Decay

The systematics of alpha-decay is being investigated with the object of finding empirical correlations between the disintegration constants for alpha-decay and such parameters as the velocity of the emitted alpha particles, the atomic number, the neutron excess, etc. So far, the most significant result has been a contribution to the understanding of the Geiger-Nuttall Rule. It is found that if the logarithm of the disintegration constant is plotted against the reciprocal of the velocity of the emitted alpha particle, for constant atomic number, straight lines are obtained. For even-even alpha-emitting nuclei the following least-squares lines are obtained:

$$Z = 84: \quad \log \lambda = 48.169 - 87.500/V$$

$$Z = 86: \quad \log \lambda = 54.050 - 99.018/V$$

$$Z = 88: \quad \log \lambda = 53.921 - 100.330/V$$

$$Z = 90: \quad \log \lambda = 51.402 - 97.405/V$$

$$Z = 92: \quad \log \lambda = 50.551 - 97.735/V$$

$$Z = 94: \quad \log \lambda = 49.181 - 97.017/V$$

These results can be compared with the following relations which have been obtained from the Gamow theory of alpha-decay:

$$Z = 84: \quad \log \lambda = 53.538 - 97.708/V$$

$$Z = 86: \quad \log \lambda = 54.819 - 100.296/V$$

$$Z = 88: \quad \log \lambda = 55.376 - 102.684/V$$

$$Z = 90: \quad \log \lambda = 56.125 - 105.072/V$$

$$Z = 92: \quad \log \lambda = 56.536 - 107.460/V$$

$$Z = 94: \quad \log \lambda = 57.072 - 109.848/V$$

In each case the empirical straight line fits the experimental data better than the theoretical line does. It is hoped that improvements in the Gamow theory may be made on the basis of this and other correlations.

If $\log \lambda$ is plotted against $1/V$ for constant values of $A-2Z$ (the neutron excess) the interpretation of the results is considerably more complicated and will be given in detail elsewhere. It appears, therefore, that the Geiger-Nuttall Rule applies to the isotopes of a given element in a much clearer way than to the members of a given radioactive series. It is also necessary to distinguish between isotopes with even Z - even N and even Z - odd N.

On the basis of the various correlations which are being studied, it should shortly be possible to make reasonably good predictions of the half-lives of such alpha-emitters as Th^{224}, Ra^{220} and Em^{216}.

A detailed summary of the work done to date will be given in a BNL report. (I. Kaplan)

Waste Disposal

In the work on permanent disposal of long-lived radioactive wastes we have determined the cation exchange capacities of certain clays as a function of cation concentration (strontium) and are developing a process for fixing the radioactive cations in the clay.

Three clays are currently under investigation in this Laboratory: a low volatile matter fullers earth (Attapulgus Clay Company) and two types of pure montmorillonite (Filtrol Corporation). In the determination of the exchange capacities, clay samples, which had been dried at 110°C, are agitated with strontium chloride solutions made from radiostrontium 89 purified from Oak Ridge Sr $(NO_3)_2$. Aliquots are withdrawn at intervals for counting. After equilibrium has been established, the clay is centrifuged, washed, and dried at 110°C. A weighed sample of the strontium saturated material so obtained is digested with concentrated HNO_3 to decompose the clay. The resulting solution is then counted. This serves as a check on the previously determined exchange capacity. The figures in Table I give the values of the exchange capacities of the clays under investigation in terms of milliequivalents of Sr per gram of oven dried clay.

TABLE I

	Capacity		
	1	2	3
Attapulgus Fullers Earth	0.31	0.32	0.30
Filtrol Montmorillonite R2532	0.48	0.46	0.46
R2472	——	0.78	0.73

1. 10 gm. clay - 225 ml. of 0.05M $SrCl_2$
2. HNO_3 extraction of saturated clay from 1
3. 0.5M $BaCl_2$ extraction of saturated clay from 1

The saturated clays are then subjected to a series of heat treatments designed to determine the amount of activity which can be fixed in clay. Thus far, temperatures in the range from 300° to 900°C have been tried for periods of about 24 hours. The samples are then treated by agitating for several weeks with a large excess of $BaCl_2$ solution. Aliquots of the supernatant solution are withdrawn at intervals for counting. Tests have shown that barium replacement is 100% complete in the case of unheated saturated clays and, therefore, this technique serves as a measure of the degree of fixation achieved by heating. Figures 1, 2, and 3 show the observed fixation as a function of temperature.

The montmorillonite minerals are known to have a crystal lattice whose c dimension is a function of the water content of the mineral. Expansion and contraction of the lattice layers is reversible if the clay is maintained at low temperatures. However, it was found by other workers that heating the clay to 400°-500°C causes a loss of swelling capacity when the clay is placed in water. Since for the most part, the exchangeable cations are located between the lattice layers, the loss of swelling ability should be associated with a lowering of the exchange capacity. This effect can be readily seen in Figure 1 and, to a lesser degree, in Figure 2 where a marked decrease in exchange capacity (or increase in per cent fixed) was observed between 400° and 500°C. Above this temperature the structure of the clay is probably altered by the loss of the elements of water from within the lattice layers and at temperatures around 800°C, the exchangeable cations appear to be fixed to a very great extent.

Future experiments will be conducted along these lines using mixed fission products, and possibly with other clays. We hope to be able to study, by means of x-ray diffraction techniques, the actual changes in the clay structure brought about by heating.

(L. P. Hatch and W. S. Ginell)

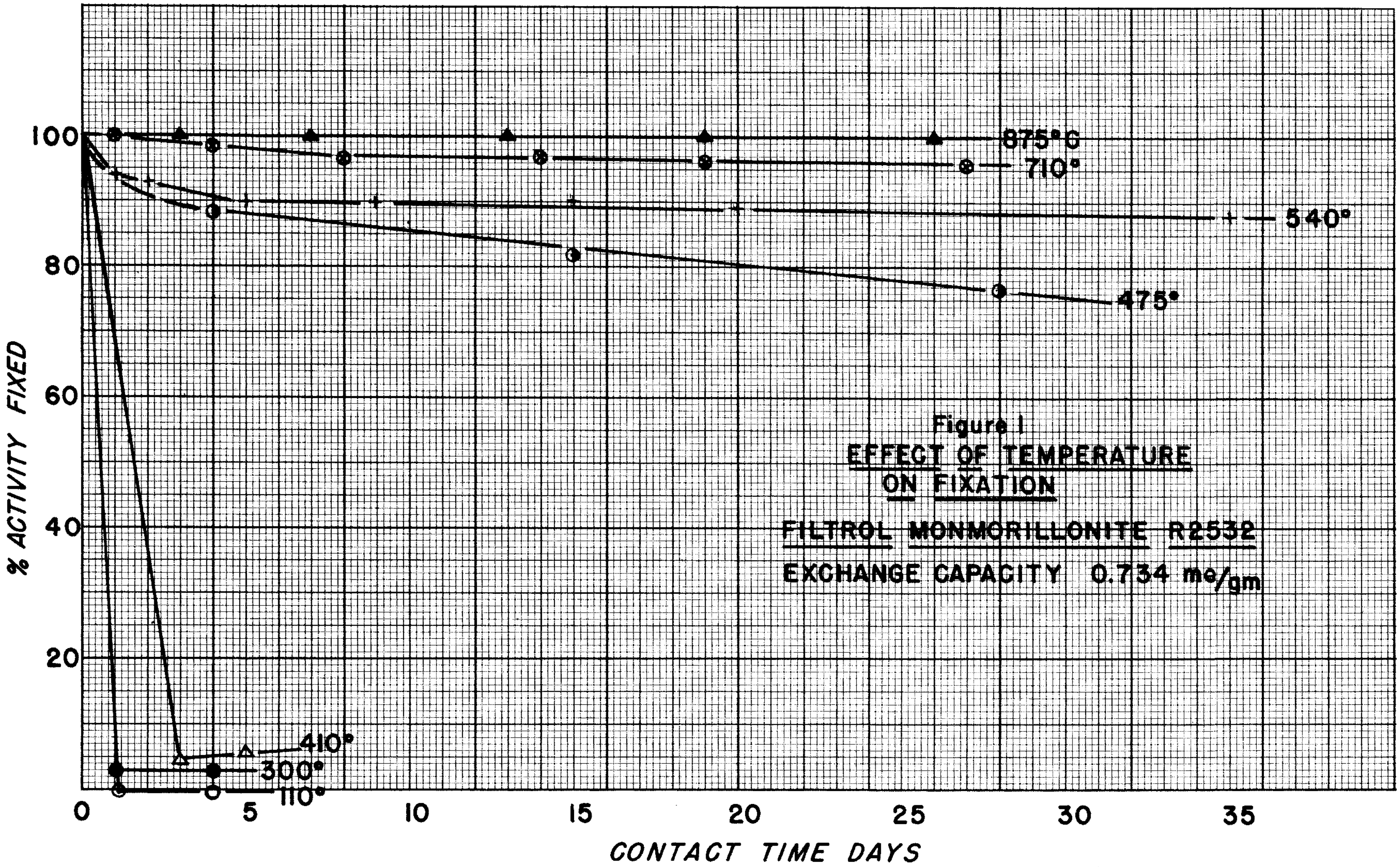

Figure 1

EFFECT OF TEMPERATURE ON FIXATION

FILTROL MONMORILLONITE R2532

EXCHANGE CAPACITY 0.734 me/gm

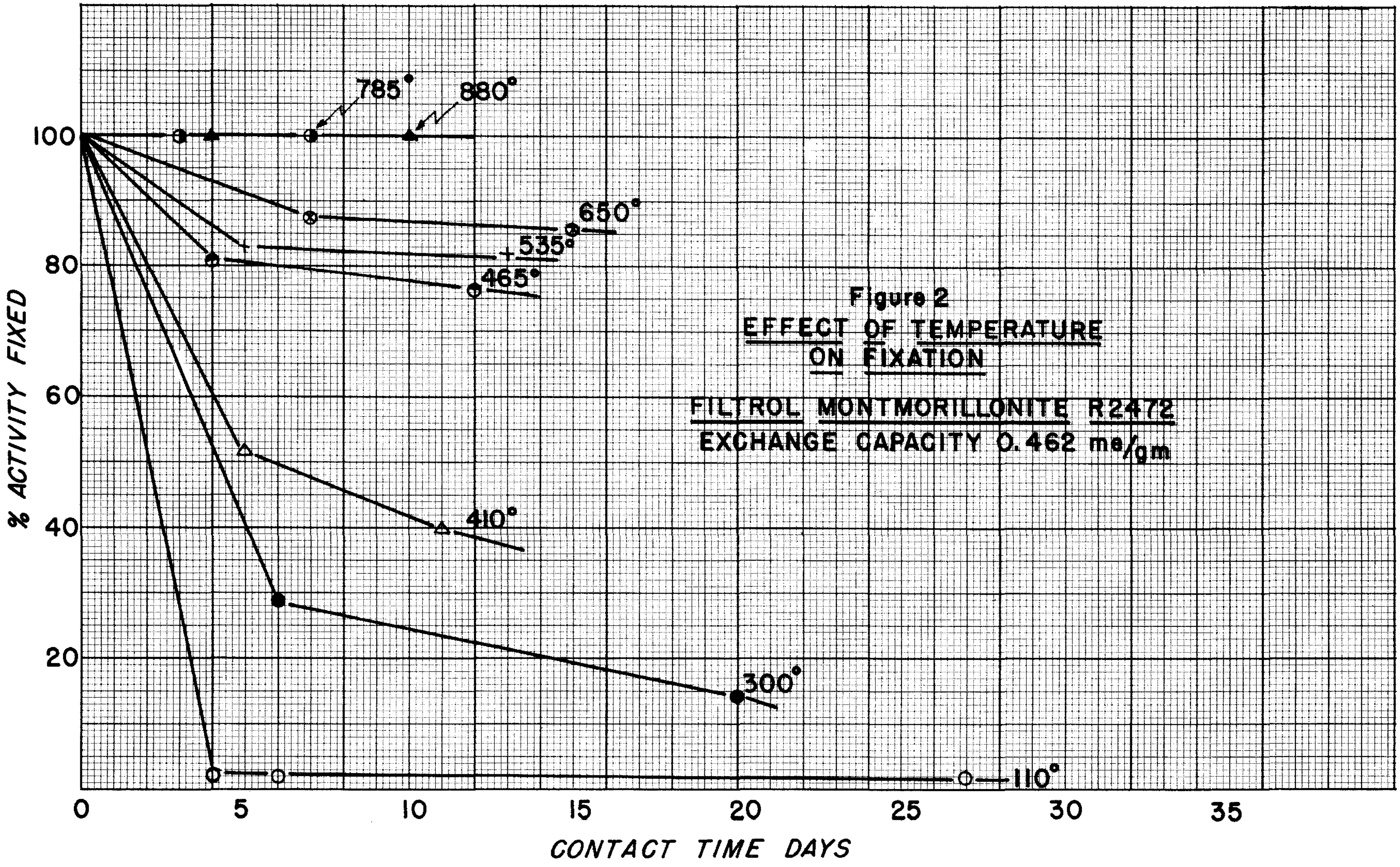

Figure 2

EFFECT OF TEMPERATURE ON FIXATION

FILTROL MONTMORILLONITE R2472

EXCHANGE CAPACITY 0.462 me/gm

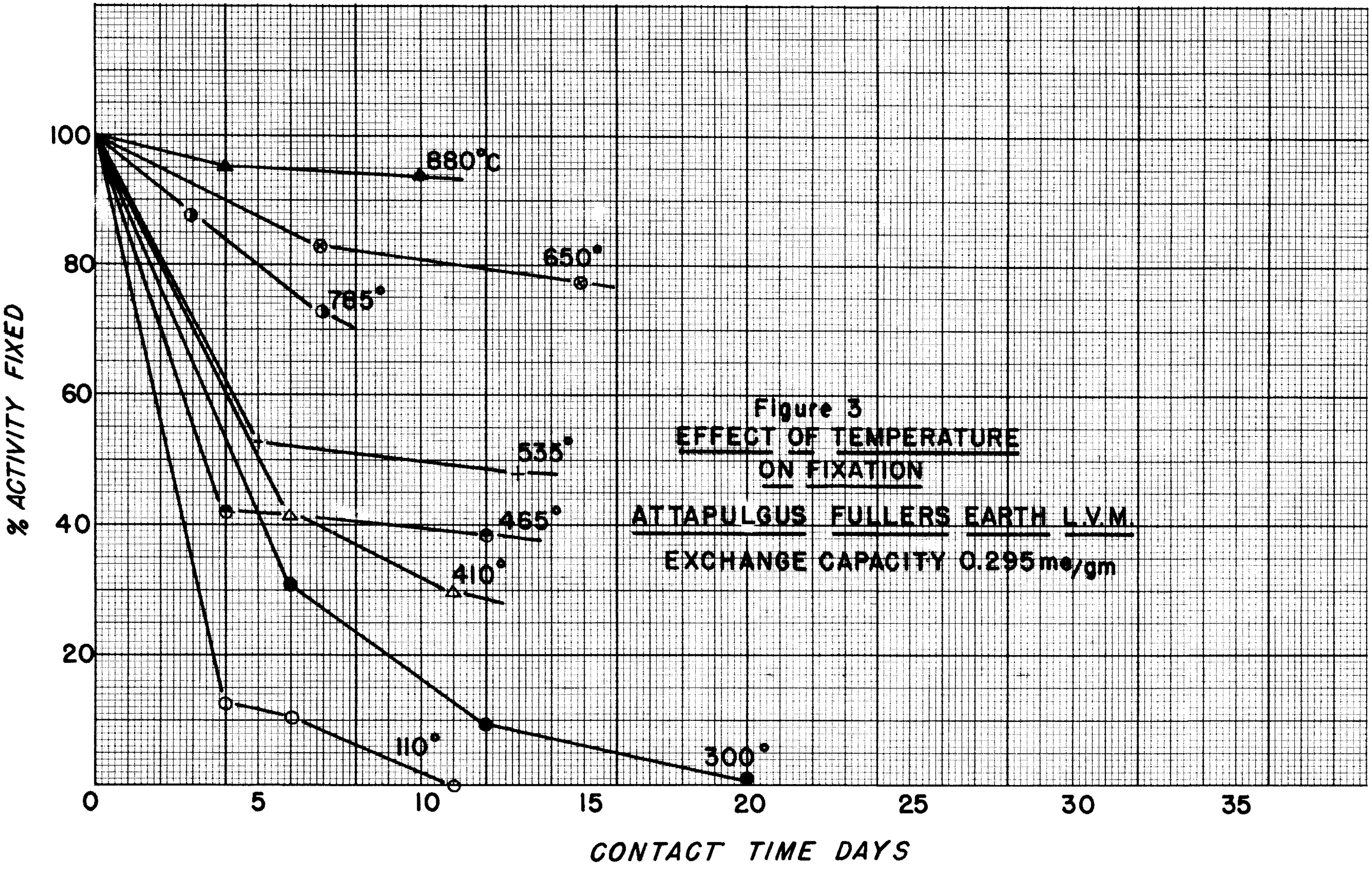

Figure 3

EFFECT OF TEMPERATURE ON FIXATION

ATTAPULGUS FULLERS EARTH L.V.M.

EXCHANGE CAPACITY 0.295 me/gm