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ORNL-1345(Del.)

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Subject Category: Particle Accelerators and High Voltage Machines

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ELECTROMAGNETIC RESEARCH DIVISION

Robert S. Livingston, Director

QUARTERLY PROGRESS REPORT, PART I
for period ending June 30, 1952

Edited by F. T. Howard

OAK RIDGE NATIONAL LABORATORY
Operated by
Carbide and Carbon Chemicals Company
A Division of Union Carbide and Carbon Corporation
Oak Ridge, Tennessee

Contract No. W-7405-eng-26

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ABSTRACT

On the 86-inch cyclotron a 41-kw beam has been calorimetered at a net ion loading efficiency of 40%; the average proton current was 1.85 ma at 22.5 Mev. Practical specific yields have been determined for (p,2n) reactions on zinc and bismuth. The investigation of products of proton-induced fission of uranium has been continued and new techniques are being used in measuring angular distribution of reaction products.

The 63-inch heavy particle cyclotron is now in operation; N^{+++} particles have been accelerated to ~ 25 Mev. In preliminary tests, induced activities have been detected in carbon, nitrogen, and oxygen targets. A hot-cathode type ion source is ready for test operation.

The 22-inch cyclotron is being used in an investigation of the problems associated with the use of rf and dc electrodes for the acceleration of protons from the ion source into the dees.

Radiation-induced corrosion in Inconel tubing containing #21 eutectic [redacted] has been produced by proton irradiation in the 86-inch cyclotron; it is shown that the corrosion was not due to thermal effects.

Approximately 150 grams of highly purified U 238 ($<$ 5 ppm U 235) have been prepared, and two grams of thorium 230 (ionium) has been enriched to 90.6%.

PREVIOUS QUARTERLY REPORTS

Quarterly reports have been issued by the Electromagnetic Research Division for the periods ending:

September 30, 1949	Y-495
December 31, 1949	Y-548
March 31, 1950	Y-584
June 30, 1950	Y-637
September 30, 1950	Y-676
December 31, 1950	Y-722
March 31, 1951	Y-767
June 30, 1951	Y-795
September 30, 1951, Part I	ORNL-1173
December 31, 1951, Part I	ORNL-1269
March 31, 1952	ORNL-1339

REPORTS PREPARED DURING THE PAST QUARTER

Harmatz, B., McCurdy, H. C., Case, F. N., and Livingston, R. S., Enrichment of Uranium 236, ORNL-1169.

Livingston, R. S. and Boch, A. L., The 86-Inch Cyclotron, ORNL-1196.

Livingston, R. S., Martin, J. A., and Murray, R. L., The Cyclorator: A Device for the Separation of Isotopes of Time of Flight in a Magnetic Field, ORNL-1309.

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THE 86-INCH CYCLOTRON

The operation of the 86-inch cyclotron has improved steadily in both continuity and quality of performance during the quarter. Earlier in the period some modifications were made to provide the increases in power and voltage required to attain the increased energy made available by modifications completed during the preceding quarter. Energy measurements by foil stack, by calorimetry, and by production yields indicate the average energy of the beam is now approximately 23 Mev at 30.5 inches. This value is well established, since many runs have been made at this radius.

Cyclotron Performance

The period of adjustment and development following the shutdown of the previous period continued into the present quarter. In these tests for adjusting the machine and determining its performance, grazing-incidence type aluminum targets were used because of the high beam intensities they withstand. Profile runs were made for determining the shape and position of the beam, and stack-foil target runs for measuring the energy of the beam.

As the performance of the machine improved, more time was spent on isotope production, physics, and radiation damage runs; for the last half of the period, the work was almost entirely of this nature. In June, two isotope production runs were made of 36,000 and 49,000 microampere-hours, with a production yield of 90% or better. In addition, there were several runs averaging 20,000 microampere-hours as well as numerous runs for physics, radiation damage, and production of Ga 67 where shorter bombardments were desired.

A new record proton beam current was measured on a 6" by 10" aluminum target of the grazing-incidence type. With the cyclotron operating steadily at 0.5 ma and at 22.5 Mev, the beam was increased progressively until the metered beam current approached 2 ma. At each level the power dissipated on the target was measured calorimeterically. The maximum beam power measured in this manner was over 41 kw, corresponding to an average beam current of 1.85 ma. The power input to the oscillator was 330 kw and the net ion-loading efficiency was 40%, that is, 40% of the power expended in acceleration of ions was to the target. There was a two-fold increase in electrical efficiency as the beam was increased from 0.5 ma to 1.85 ma.

A 750 μa-hr bombardment of zinc is scheduled each week for the production of Ga 67 used in cancer research by the Oak Ridge Institute of Nuclear Studies.

A summary of the bombardments made during the months of April, May, and June follows:

Experimental	41
Isotope Production	31
Physics	72
Radiation Damage	16
Total	160

Construction and Revisions

A 42-inch shielding wall of concrete block has been installed at the target handling area in the cyclotron pit. A viewing window, also 42 inches thick, is a clear plastic chamber which will be filled with zinc bromide. By means of a manipulator extending through the wall, an operator is able to remove a radioactive target and deposit it in a lead-shielded container.

Carbon lips of a new design were installed on the edges of the dees to reduce sparking at the increased dee-to-dee voltage, 400-500 kv, required for operation at the high energy level.

After local interference with television reception had been reported all components that project from the cyclotron tank were shielded. A signal can still be detected in the cyclotron building but those residents of Oak Ridge whose TV antennae are directed towards Y-12 get no interference.

An arrangement for filling the liquid nitrogen traps from outside the cyclotron shielding wall and a guage for indicating liquid level have been installed. The liquid nitrogen traps can now be filled while the machine is in operation, without exposing personnel to radiation.

In the past, the ion source was adjusted manually for best operation; no adjustment was possible while the machine was in operation. A rotating device, remotely controlled by Selsyn motors, now permits the source to be rotated slowly to optimize the source position while the machine is in operation.

THE 63-INCH CYCLOTRON

The heavy particle cyclotron is now in operation; the first beam was obtained on May 20. With a brass target at the 21" radius, the gamma count at the top of the cyclotron tank was measured to be eight times background. Since the singly-ionized nitrogen ions at this radius have an energy of only 2 1/2 Mev, it may be concluded that the burst of radiation observed was indeed due to triply-charged nitrogen ions impinging on the target. Preliminary activation experiments have been performed with the internal beam of the cyclotron.

Performance

The performance of the rf system of the cyclotron has been highly satisfactory. There has been no difficulty in obtaining oscillations in the resonant system at the proper frequency, or in raising the dee voltage to its expected value. The cyclotron now operates with about 20 kw power input. The efficiency of the oscillator is approximately 50% and the dee-to-ground voltage can be raised to as high as 50 kv without sparking inside the vacuum system. A negative voltage bias, 1 to 2 kv, is applied to the dees in order to sweep out any ions that may be formed while oscillation is being established. The deflector which will be used for the extraction of the beam has been tested and found to hold 50 kv negative potential. The electrical field strength at the entrance to the deflector gap is between 50 and 60 kv/cm.

The arc source used for the initial tests is of the hollow-anode type. Two water-cooled tantalum cathodes, at approximately 5 kv negative potential, are inserted on opposite ends of a hollow carbon anode which is at ground potential. Gas is fed through a tantalum-tipped tube directly into the arc chamber. Under operating conditions the arc voltage is approximately 600 volts,

the current is around 2 1/2 amperes, and the flow of nitrogen gas into the arc is less than 1 cc/min. Dc tests of this type of source indicate that the ratio of N^{+++} to N^{+} is approximately 1:1. The alignment of the source with the magnetic field is extremely critical, as was expected, and makes it necessary to provide for external adjustments of the ion source.

In attempts to determine the beam current at various radii the following probe currents were measured:

Beam Radius (inches)	Probe Current (μa)
5	2000
10	500
14	170
18.5	30

Current measurements beyond 18.5" were unreliable; the current at the maximum radius, 25.5", is estimated to be of the order of one microampere. Gamma radiation produced by the triply-charged nitrogen ions is approximately 400 times background, measured at the top of the cyclotron. The total number of neutrons produced is approximately 10^5 per second.

Preliminary Activation Experiments

To make sure that the beam in the cyclotron was indeed of high energy several targets were bombarded with the internal beam at a radius of 24.5". A carbon (graphite) target gave rise to 112-minute and 15-hour activities which are assigned to F 18 and Na 24 respectively. No other activity was observed with a half life between one minute and one day. Oxygen, bombarded in the form of CuO, showed a 2.5-minute activity and a 112-minute activity. The 2.5-minute activity is due to either Al 28 or P 30, but since formation of P 30 would be a radiative capture reaction, which is highly unlikely, the short activity is assigned to Al 28.

When nitrogen was bombarded in the form of TaN, 2-minute, 10-minute, 112-minute, and 15-hour activities were observed. In these first experiments, the amount of oxygen or carbon contamination in the TaN target was not known, hence it is difficult to assign any but the 10-minute activity as unequivocally due to N 13 formed by the reaction $N\ 14 + N\ 14 \rightarrow N\ 13 + N\ 15$. Considerably more work is necessary before any conclusions may be reached regarding nuclear reactions involving high energy nitrogen ions impinging on these light elements. The work planned includes a verification of the beam energy and identification of the reaction products by means of radiochemical techniques, as well as by beta ray spectroscopy.

THE 22-INCH CYCLOTRON

The 22-inch cyclotron is currently being used in investigations of the problems associated with the use of rf and dc electrodes for the acceleration of protons from the ion source into the dees. Radio frequency accelerating electrodes of three geometries mounted on the lower dee near the ion source have been examined for increased proton current. An electrode curved to focus ions towards the median plane gave a 15 percent greater output than any other geometry examined. Optimum geometry for the rf accelerating electrode has not yet been determined.

Calculations of the possible advantage of using dc potential on the accelerating electrode to obtain high intensity beam currents indicated that the 22-inch cyclotron might be used effectively in the study of dc injection of ions into the dees. A dc accelerating electrode was mounted on a steatite insulator from the liner on the source-anode side. The mountings provide a rotation adjustment in the vertical plane perpendicular to the dee edge. Since the ion source has translation adjustments in the horizontal and the vertical planes and a z-wise rotation in the vertical plane perpendicular to the dees, adequate relative positioning of source, electrode, and dees was available. A modification was made on the z-wise rotation adjustment of the source and a five-segmented water-cooled target probe for measuring the z-wise beam distribution has been fabricated. Data which have been obtained in the dc injection experiments are currently being studied.

NUCLEAR PHYSICS RESEARCH

Both the primary protons and the secondary particles resulting from proton bombardment in the 86-inch cyclotron are being used for investigations of nuclear reactions. The practical specific yields for the reactions Zn 68(p,2n)Ga 67 and Bi 209(p,2n)Po 208 have been measured and will be reported elsewhere. [redacted] An experimental search for the dineutron has been completed and will be reported in ORNL-1382 and ORNL-1383. The half life of potassium 44, from Cu 44(n,p)K 44, measured ~ 22 minutes; other properties of the isotope are being studied. The identification and study of the properties of the isotope boron 13, produced by C 13(n,p)B 13, has been undertaken. The investigation of proton-induced fission has been continued and new techniques have been developed for measuring the angular distributions of reaction products.

Proton-Induced Fission

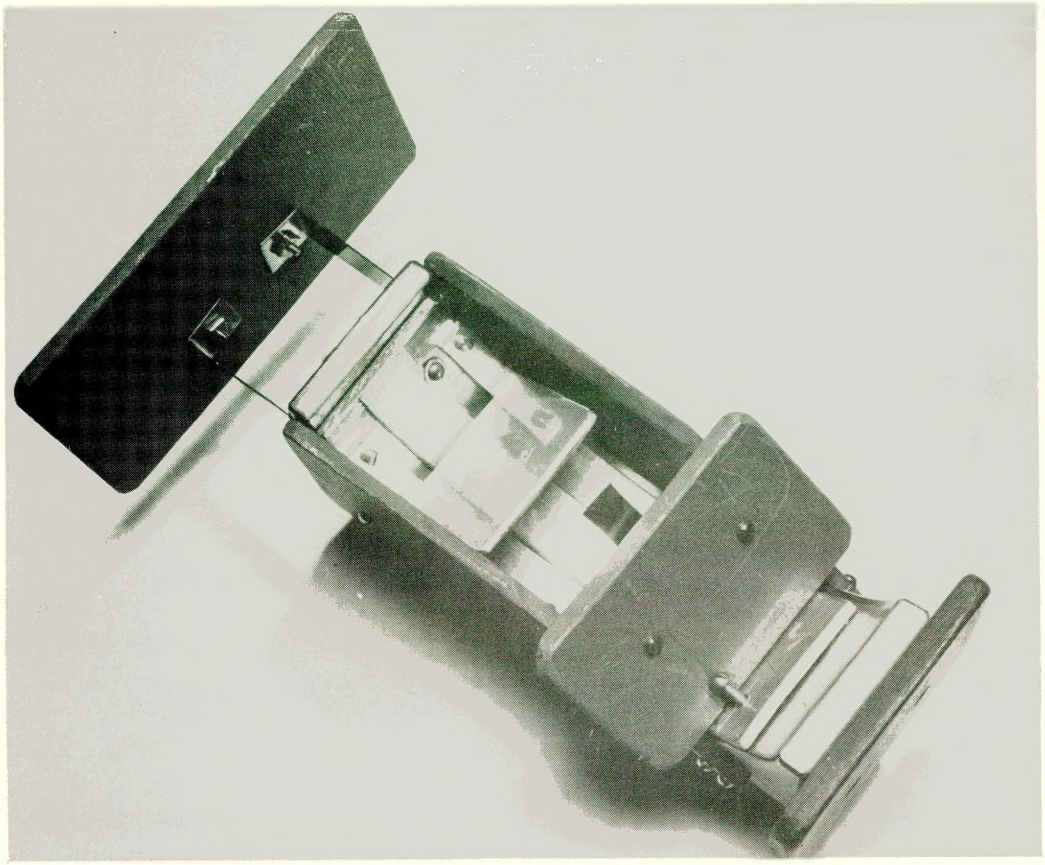
The study of fission product yield ratios as a function of the energy of protons on uranium has been continued. In preparation for further experiments a number of fresh stock solutions have been prepared and carrier solutions standardized. Resin columns and other specialized equipment have been installed to facilitate specific operation in the chemical analyses. With this equipment it is possible to separate Ag, As, Ba, Cd, Cu, Mo, Nd, Pr, Ru, Sn, and Y. In initial tests four single uranium foils and one foil stack have been bombarded and chemically analyzed.

Angular Distributions

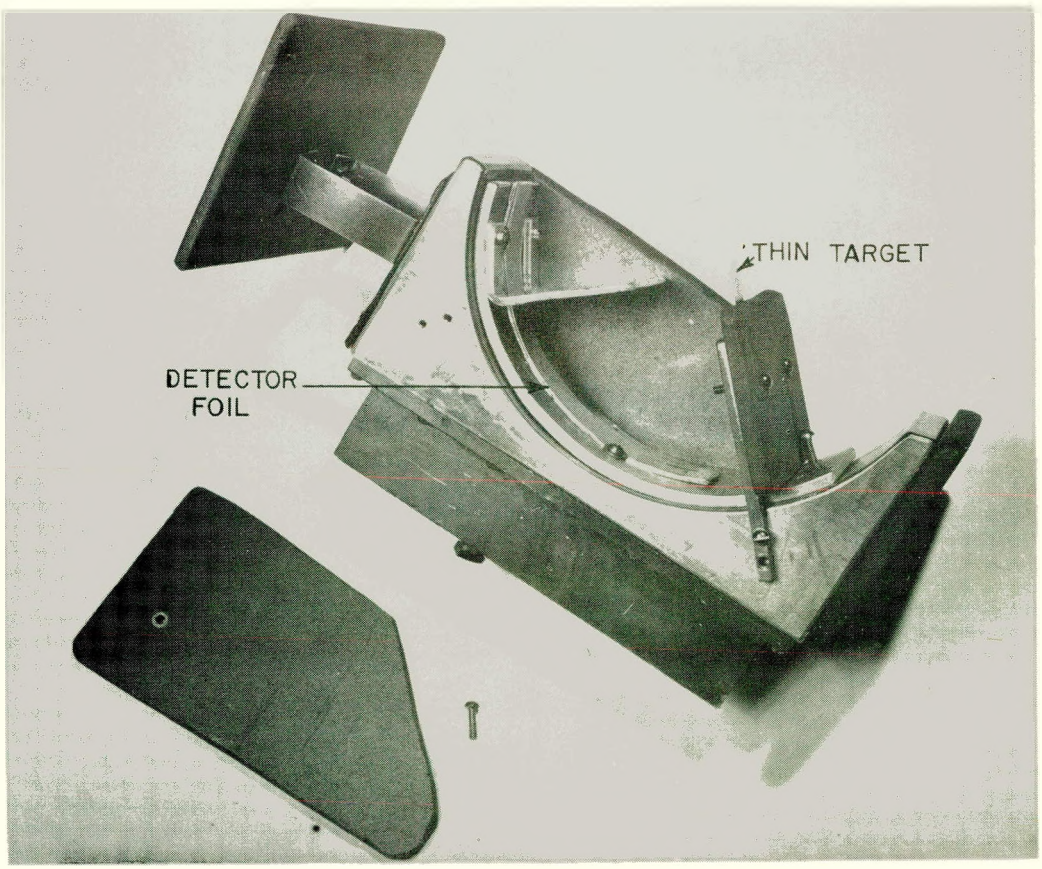
A target to be used with the internal beam of the cyclotron has been designed to measure angular distribution, Figure 1. The new target was constructed after various arrangements of activation detectors and graphite shields had been tested.

The first project for which the target is being used is the measurement of angular distribution of tritons from the reaction Be 9(p,t)Be 7. The tritons are detected by measuring the 1.6-hour activity of Co 61 from the reaction Co 59(t,p)Co 61. The Be 7 possesses no excited states between 0.4 and 7 Mev above the ground state; consequently, the tritons are essentially monoenergetic with their energy varying with observation angle (due to center-of-mass effects) from 0 to 10 Mev. Since the detection efficiency of cobalt is a function of energy, the detecting foil is covered with sufficient absorber thickness to reduce the energy at each angle to a standard energy of 6 Mev so that the distribution can only be measured up to about 90° (center-of-mass system). A separate experiment, not using absorbers, is also underway to determine the detection efficiency of cobalt, which is essentially the excitation function for the (t,p) reaction. The preliminary results of the first experiment indicate isotropy, within a factor of two, with no regular variation. The second experiment indicates that the cross section rises steeply at an energy about equal to the coulomb barrier energy, and then flattens.

Methods of detecting elastic protons for the purpose of angular distribution measurements were studied. The most suitable method found employs the 38-minute activity of Zn 63 from Cu 63(p,n)Zn 63. When the detecting foils are covered with a suitable aluminum absorber, the detection efficiency as a



A. ASSEMBLED



B. SIDE PLATE REMOVED

FIGURE 1. ANGULAR DISTRIBUTION TARGET

function of proton energy can be made to rise very steeply at any desired energy (chosen high enough to eliminate inelastic protons). The angular distribution of protons elastically scattered from beryllium has been measured. It drops off quite rapidly with increasing angle, and up to 100° shows no secondary maximum--as might be expected from diffraction effects.

Methods of detecting deuterons were also studied. The reactions Pb 208(d,p)Pb 209 (3.3 hr) and Mg 26(d,p)Mg 27 (10 min) were found to be difficult for routine use due to the necessity for accurate determination of long-lived activities that must be subtracted off. Radiochemical techniques for the lead reaction are now being tried.

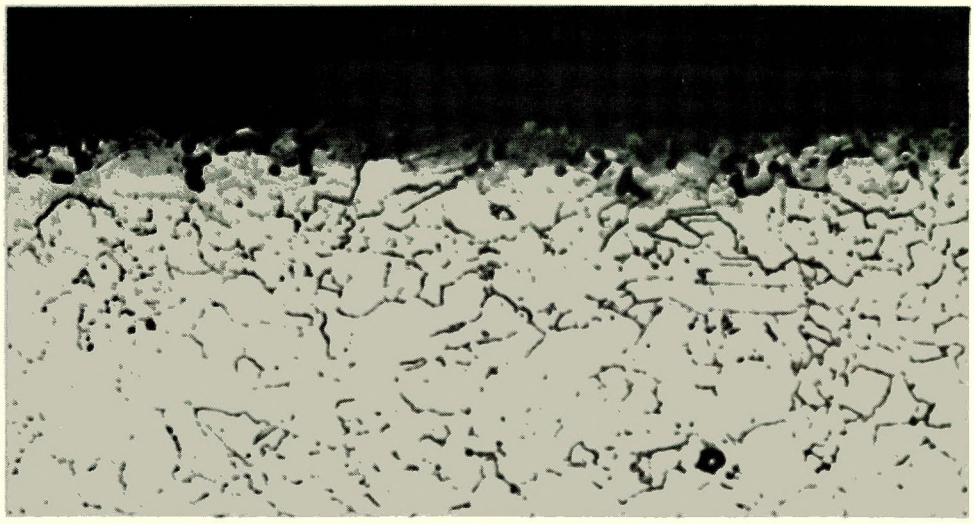
RADIATION DAMAGE

Reactor Fuel Element Corrosion Under Proton Irradiation

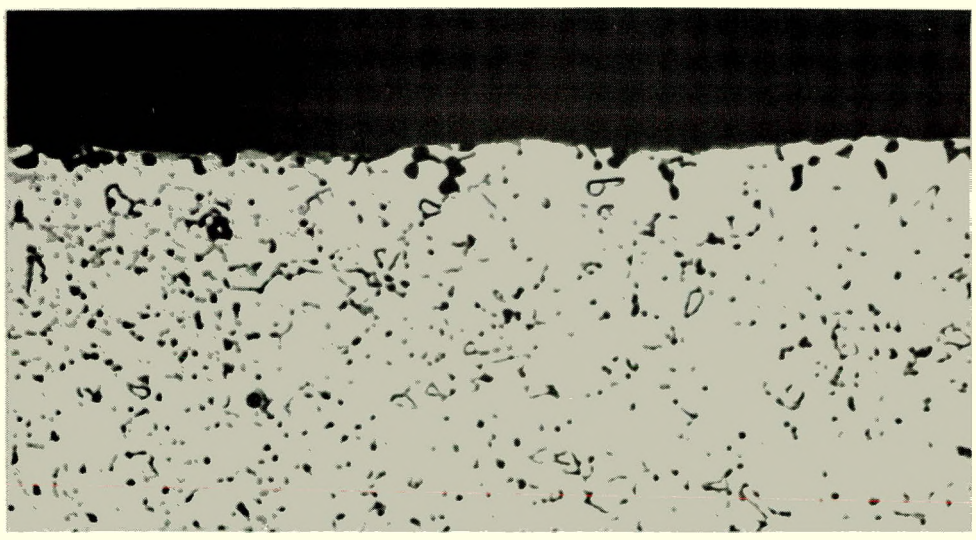
Bombardment of liquid eutectic fuels in Inconel has been resumed since the 86-inch cyclotron is again in operation. The higher beam energy, $\sim$ 23 Mev, now obtained on the cyclotron is expected to have little effect on the type of damage incurred by the bombarded fuel elements.

Bombardments of less than eight hours duration at 815°C had previously produced less than 0.0005" of corrosion, which is the limit of detection with present methods. A single eight-hour run, however, showed some corrosion (0.025") in Inconel at the #14 eutectic-Inconel interface. Several eight-hour bombardments of #21 eutectic were then made for comparison with earlier results. The #21 eutectic has the following composition: NaF, 4.8 mole %; KF, 50.1 mole %; ZrF, 41.3 mole %; and UF_4, 3.8 mole %.

Of the three eight-hour bombardments thus far completed with #21 eutectic chemical and metallurgical results have been completed in only one case. Chemical analysis of the eutectic for trace quantities of Inconel elemental components indicated only minor increases in the bombarded specimen, but microscopic metallurgical examination showed significantly increased corrosion in the bombarded area of the Inconel tubing, Figure 2a. As a control a second Inconel tube, Figure 2b, was filled with the same batch of eutectic and subjected to the same thermal history, 815°C, in a vacuum furnace for eight hours. The net effect of the bombardment with 23 Mev protons at a power density of about 2500 watts/cc has been to increase the depth of corrosion by 0.0015". It seems safe to conclude that corrosion is enhanced by proton bombardment, although perhaps not at a rate which is excessive for engineering application. More quantitative work is needed, however, to answer this question unequivocally.



a. PROTON IRRADIATED



b. CONTROL

FIGURE 2. RADIATION-INDUCED CORROSION

[redacted] It was possible to concentrate somewhat more upon several of the systematic problems inherent in the cyclotron experiments. These are discussed in the following two sections.

Thermal Gradient Across Bombarded Fuel Element

During the actual proton bombardment, the temperature of the bombarded side of the fuel element target is controlled and held at 815°C. To dissipate the beam energy it is necessary to mount the target on a water-cooled copper tube. As a consequence the thermal gradient across the tube is quite steep, the boundary temperatures being 815°C and 10°C. Since control samples in the uniformly heated furnace have no such gradient, it is possible that the enhanced corrosion observed in the bombarded targets may be a mass transfer of some sort depending primarily upon this thermal gradient.

To examine this possibility, an Inconel fuel element was mounted on a water-cooled copper tube and the whole assembly was placed in the control furnace. Under these conditions the boundary temperatures were 815°C and 40°C. This specimen was held at this temperature gradient for eight hours so that it also served as a control for the bombardment shown in Figure 2a. Although there is no assurance that the nature of the gradient function, aside from boundary conditions, is the same, no evidence of the mass transfer type of corrosion due solely to a thermal gradient is found. The enhanced corrosion observed in Figure 2a seems to be quite definitely associated with the proton irradiation.

It is also of interest to note that, by the nature of these experiments, the corrosion detected is primarily in a region where most of the proton energy is dissipated by ionization. Chemical aspects of corrosion may be expected to be sensitive to ionization, while corrosion dependent upon micro-diffusion will occur primarily in the region of knocked-on atoms.

Knock-Ons and Ionization

In the first part of its path through matter an energetic charged particle, such as a proton, loses energy by ionization and excitation of the adjacent atoms; at lower energy, near the end of its range, the energy exchange is by the knock-on process, in which the atoms of the absorber may be displaced from their lattice sites both by the primary particle and by the recoil atoms. Generally, too, the number of secondaries produced by an energetic light primary particle is very large. Since radiation damage, as determined by measurement of mechanical properties and micro-diffusion rates, is due to displaced atoms while certain chemical effects, such as dissociation of water and coloration effects, are due to ionization, it is of some interest to try to separate these factors in corrosion. If, for example, it can be shown that corrosion is enhanced primarily by knock-ons, then the problem of the initiating particle is lessened, since in the last analysis the damage will be caused by the large number of recoil atoms in the target material itself.

An experiment designed to separate these effects is being initiated. For a proton the transition between excitation-ionization and knock-on occurs at about 10 kev, that is, above 10 kev the particle loses most of its energy by ionization, below, mostly by knock-on. Hence, a simple absorbing wedge

inserted in the proton path ahead of the target would result in protons of all energies from 22.5 Mev to zero energy striking the target. A simple metallurgical examination of the target for corrosion as a function of position on the target face should determine whether low energy protons (knock-ons) or high energy protons (ionizations) has the greater effect on corrosion rate.

As an alternate approach to the same problem, a preliminary survey has been made of the feasibility of mounting a rotating disk of continuously varying thickness in the path of the proton beam. This system would employ a graphite disk of wedge-shaped cross section, the peripheral area of which would attenuate the beam energy and thus produce a uniform distribution of knock-ons through the target. Such an energy modulator would prove very useful in many types of radiation damage studies where the phenomena induced depend upon displaced atoms and in which a uniform distribution of damage in a target is advantageous. It may be, however, that the application of the rotating absorber to these problems will be delayed until the proton beam of the 86-inch cyclotron is deflected.

SPECIAL SEPARATIONS

Facilities for Plutonium Separation

Electromagnetic facilities to be used in plutonium enrichment were pre-tested by a one-stage separation of U 238 from uranium stock which had been depleted in U 235. The vacuum tank and the related vacuum systems had been modified to confine dispersed material to a smaller volume. These changes had included reducing the area of the manifold opening to the tank by a factor of three and isolating the tank from the roughing vacuum system. In this test operation the operating pressure was found to be adequate. The gloved box in the wash area was used in disassembling the collector and for recovering the product by vacuum scraping the U 238 collector pocket. The product has not yet been assayed but it is expected to total 150 grams of U 238, containing not more than 5 ppm of U 235.

The wash area and laboratory facilities for removal of unresolved uranium deposits on equipment and for extraction of wash solutions are nearly ready.

Enrichment of Thorium 230 (Ionium)

The thorium isotope Th 230 has been enriched from a 294 gram supply of thorium containing 10.1% Th 230 (ionium). The thorium oxide was heated to 450°C for five hours in a stream of phosgene for conversion to the tetrachloride. This charge was then run in a two-arc ion source capable of operating at temperatures up to 850°C. The Th^+ ion beam was baffled to a 10° angle (+6°, -4°) and collected through 1/16" wide collector slots.

Two grams of thorium oxide were separated in which the desired isotope, Th 230, was enriched to 90.6%. This corresponds to a two-mass-unit enhancement factor of 87. Additional thorium separations scheduled will require recycling the starting feed.

Shipments of Enriched Uranium Isotopes

Distribution of electromagnetically separated uranium isotopes to Atomic Energy Commission laboratories during April, May and June is tabulated below:

Oak Ridge National Laboratory, X-10	218 mg U	96.7% U 236	
Oak Ridge National Laboratory, X-10	11 mg U	95.3% U 236	
Naval Research Laboratory	20 mg U	99.9993% U 238	
Oak Ridge National Laboratory, X-10	10,000 mg U	99.9993% U 238	
University of California Radiation Laboratory	1,000 mg U	99.9993% U 238	
Oak Ridge National Laboratory, Y-12	990 mg U	99.9956% U 238	
Knolls Atomic Power Laboratory	1,000 mg U	99.9956% U 238	
Chalk River, Canada	36,786 mg U	99.9947% U 238	
Naval Research Laboratory	20 mg U	99.945% U 238	
Oak Ridge National Laboratory, Y-12	1,005 mg U	99.945% U 238	
Oak Ridge National Laboratory, X-10	10 mg U	99.994% U 235	
University of California Radiation Laboratory	502 mg U	99.994% U 235	
Oak Ridge National Laboratory, Y-12	1,002 mg U	99.79% U 235	
Oak Ridge National Laboratory, Y-12	1,500 mg U	99.74% U 235	
Los Alamos Scientific Laboratory	3 mg U	94.00% U 234	
General Electric Company - Hanford	1,693 mg U	20.18% U 234	

ELECTROMAGNETIC FUNDAMENTALS

Ion Source for 63-Inch Cyclotron

The preliminary testing of a hot-cathode ion source for the 63-inch cyclotron is now complete and the source is ready for actual tests in the cyclotron. In dc tests the output of the source, measured after the beam had passed through a 180° path in the magnetic field, was: 8 ma of N^{+}, 2 ma of N^{++}, and 2 ma of N^{+++}. The N^{+++} beam was scanned and the peak found to be six times background. The expected filament life is in excess of ten hours.

DC Injection

Steady operation of the dc injector ion source at 100 volts and 8 to 10 amps was achieved by opening the defining slot to 1/8" by 1/2". The maximum proton output obtainable with this source, for 15 kv dee-to-ground potential, was only 30 ma, as compared with 75 ma previously obtained from the original source with a smaller defining slot. In the dc test the new source output was 180 ma. It was never possible to make a dc test without high drain to the accelerating electrode. This is a definite clue to the lower output obtained in the rf tests in which the drain to the accelerating electrode was also high. It appears that for steady high output with rf on the dees further design work is necessary.