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ORNL-1269

ELECTROMAGNETIC RESEARCH DIVISION

QUARTERLY PROGRESS REPORT

Part I

FOR PERIOD ENDING DECEMBER 31, 1951

AEC RESEARCH AND DEVELOPMENT REPORT

OAK RIDGE NATIONAL LABORATORY
Y-12 AREA
OPERATED BY
CARBIDE AND CARBON CHEMICALS COMPANY
A DIVISION OF UNION CARBIDE AND CARBON CORPORATION
OAK RIDGE, TENNESSEE

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Subject Category: Particle Accelerators and High-Voltage Machines

ELECTROMAGNETIC RESEARCH DIVISION
Robert S. Livingston, Director

QUARTERLY PROGRESS REPORT, PART I
for Period Ending December 31, 1951

Edited by F. T. Howard

January 15, 1952

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-7205-eng-26

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Particle Accelerators and
High-Voltage Machines

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ABSTRACT

Operation of the 86-inch cyclotron has been continuous except for a three-day interruption caused by a water leak inside the vacuum chamber; monthly output has averaged over 210,000 μa-hr; the average continuous (168-hour week) beam power has been approximately 5.5 kw; and the energy of the beam has been determined to be $\sim$ 19.4 Mev.

No changes due to radiation damage have been detected in Inconel tubes containing UF_4-NaF-KF eutectic which were irradiated in the 86-inch cyclotron with power inputs up to $\sim$ 415 watts/cc.

The major components of the 63-inch cyclotron have been fabricated and are being tested as the machine is being assembled. On the 22-inch cyclotron a radius-wise examination of the fine structure of the proton beam reveals definite proton orbits.

Several grams of very pure uranium 235 have been prepared; the chemical recovery for first-stage separation of U 236 has been established as 97%; and facilities for laboratory-scale separation of plutonium isotopes are being prepared.

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THE 86-INCH CYCLOTRON

Operation of the 86-inch cyclotron has continued at a high level, the output averaging over 210,000 μa-hr per month. For a 7-day-week, 24-hour-day schedule this represents an average continuous beam power of approximately 5.5 kw. Full time operation has permitted only minor alterations; a shutdown for modifications is scheduled for January, 1952. A number of factors have been investigated in order to improve and better define cyclotron performance and to improve control of radiation hazards.

CYCLOTRON OPERATION

Operation of the 86-inch cyclotron has continued on a full time basis of 168 scheduled hours per week with 144 hours per week assigned to isotope production and the remaining 24 hours available for physics and radiation damage bombardments. The average weekly beam innage, beam-on-target time, fluctuated from 25% to 88%. The latter is the highest innage recorded to date for a seven-day period of operation. A shutdown of approximately four weeks is scheduled for January 1952 to permit several needed modifications of the equipment.

Cyclotron Performance

The cyclotron has been in continuous operation except for a three-day interruption due to a water leak which developed in the header of the dee cooling circuits. It was necessary to remove the top of the vacuum tank to make repairs. While several minor vacuum leaks were encountered during the period, this is the first major difficulty experienced in the past six months.

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During the past quarter the beam innage and the microampere-hours of beam on the target were as follows:

Month	Innage (%)	Total Beam on Target (μa-hr)
October	65	260,000
November	55	187,500
December	55	203,500

The present dee system has now had a total of 1,063,000 microampere-hours of operation and at the last inspection all components were still in good condition. No further trouble has been experienced with tank heating since the water-cooled liner was extended upward around the dee support frame. Due to operation commitments, no time was allotted for further measurements of the maximum beam currents obtainable from the cyclotron; several high current targets are being fabricated, however, for future use.

The total of 159 cyclotron bombardments completed during the quarter are tabulated as follows:

Isotope Production	37
Experimental	3
Radiation Damage	48
Nuclear Physics	71
Total	159

Modifications

Changes in cyclotron components were held to a minimum because of reluctance to interrupt operation commitments, and because a scheduled shutdown was planned for January, 1952. Modifications attempted were as follows:

1. The use of a copper-plated stainless steel ion source stem
2. Modification and relocation of the water connections to target

Ion Source Tube. The ion source is mounted on a long stem consisting of a two-inch diameter copper pipe approximately 14 feet long. Extreme care is necessary to prevent bending the soft copper tube during the installation and removal operations. Any distortion in the tube results in added difficulties in the proper positioning of the ion source. A more rigid stem was fabricated from copper-plated stainless steel tube. Although it was not so easily bent, this tube was unsatisfactory. The copper plating tended to bubble and pit when subjected to the radio frequency and the tube evidently heated unevenly during operation, resulting in a serious misalignment of the ion source and accelerating slit. Other methods are being studied for maintaining the proper alignment.

Target Water Connections. The time required to detach water hoses from the target dolly necessitated an undesirable exposure of operation personnel. In order to reduce this radiation exposure, quick disconnect couplings have been installed which enable the operator to remove both water lines in less than twenty seconds. With longer hoses the connections have also been relocated about five feet farther away from the target position.

CYCLOTRON DEVELOPMENT TESTS

Exploration of the potentialities of the 86-inch cyclotron has continued, along with more refined measurements of performance. Potentially larger ion currents are indicated but cannot be realized until targets of greater capacity are developed. Radiation hazards associated with larger ion currents have been explored. An apparent displacement of the orbital center of the beam results in a final energy less than should be expected from $H\rho$ considerations; the causes of this displacement are being investigated.

Presence of Large Circulating Currents

Evidence of the further increases in proton current that may be possible with the present 86-inch cyclotron ion source has been demonstrated. A measurement of the dee bias drain was made for several conditions of ion source arc and of beam resonance. The leakage current was determined and the proper correction applied to the dee bias current. The results obtained were readily reproducible and are believed accurate to better than 10%. The following tabulation shows the beam current that is circulating at inner radii when the cyclotron is on resonance and slightly above resonance. The energy of the ions was estimated from oscillator loading data corrected for the measured efficiency of the oscillator. All of the readings were taken at 320 kv dee-to-dee and 1 kv dee bias.

Condition	Dee Bias Drain (ma)	Dee Bias Leakage (ma)	Ion Loading Current (ma)	Ion Loading Power (kw)	Avg. Ion Energy (Mev)
500 μa beam, on resonance	18.6	8.6	10.0	17.0	1.7
4 μa beam, above resonance	59.9	8.6	50.4	37.4	0.74

With the beam on resonance and the cyclotron operating with 500 μa on the targets, the above experiment shows that 10 ma of beam is circulating for the first few revolutions with an average energy of 1.7 Mev. With the "off resonance" condition in which almost all of the beam strikes the dees, an ion current of 50 ma at an average energy of 0.7 Mev was reached. This suggests that, when improved targets are ready, much larger beams will be obtained at the full cyclotron energy.

Focal Pattern of the Proton Beam

For determining the focal pattern of high energy protons in the beam a target was designed to hold a number of 1/32" carbon foils, Figure 1. The cyclotron beam strikes the edges of the stacked foils. After exposure to protons the foils are removed, broken into 1/2" sections (previously scored) and then the activity of each piece of foil is counted. Measurements based upon the C 12(p,pn)C 11 reaction and upon activities due to impurities in the carbon were in agreement and the results reproducible.

The distribution of activity in targets exposed under various cyclotron conditions is shown in Figure 2a. The beam shape is very sensitive to the dee-to-dee voltage and to resonance conditions, as predicted by cyclotron theory. Grounding the ion source also has a substantial effect. In all tests the beam focus was found to be off-center, about 3/4" to the west, as indicated in the typical plot in Figure 2b. When the minimum phases of the ions with respect to the electrical voltage at resonance are estimated from the radial width of the beam for given dee-to-dee voltages, results are obtained which are in good agreement with phases predicted from cyclotron theory.*

Neutron Measurements

Previous measurements had indicated the presence of an excessive neutron flux outside of the entrance maze of the cyclotron shielding wall. A wood and paraffin door designed to reduce the neutron flux has now been installed and neutron measurements indicate an attenuation factor of 200 straight through the door, but there is considerable leakage around the edges. The

* Cohen, B. L., Fixed Frequency Cyclotron Theory, April 4, 1951, Y-757.

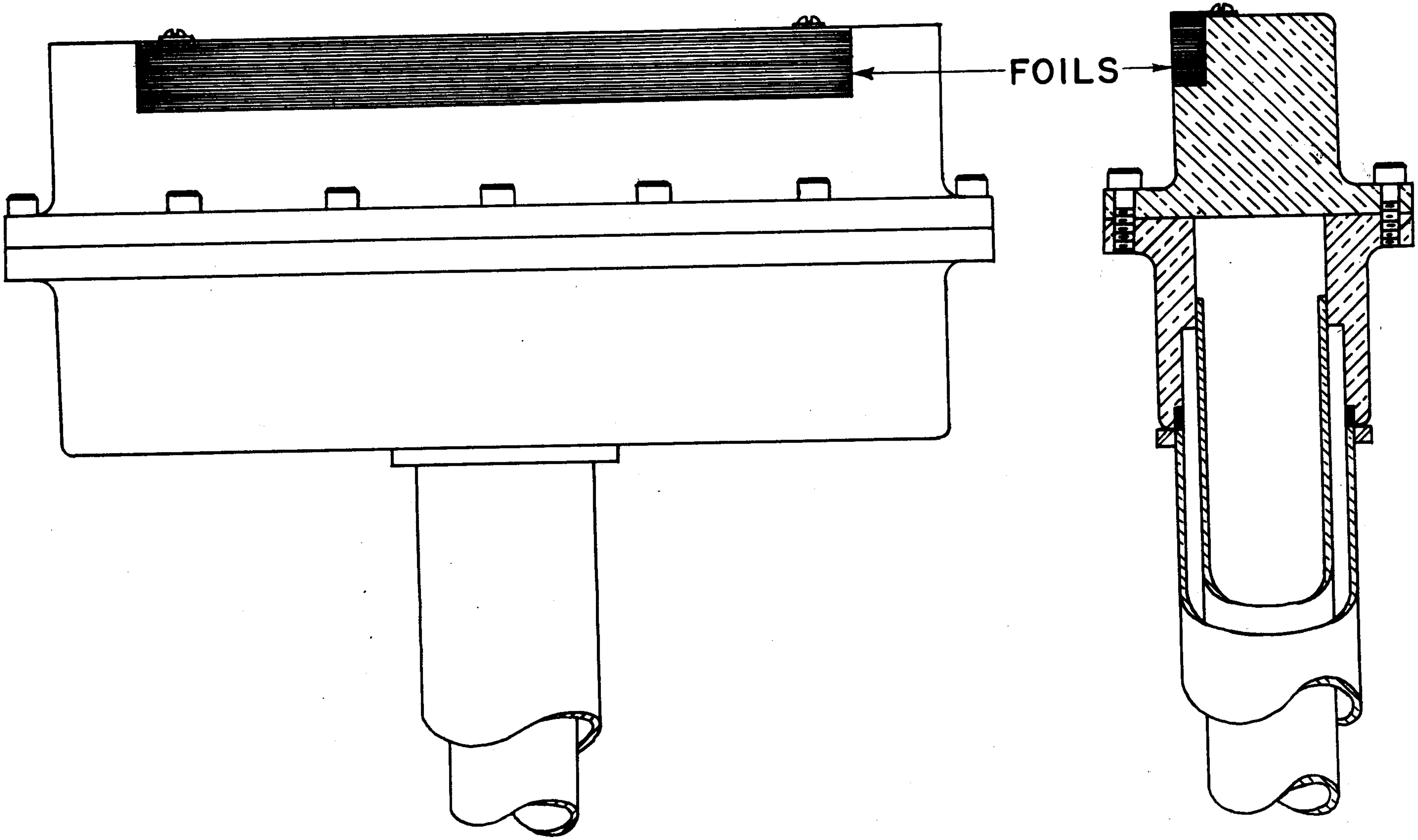
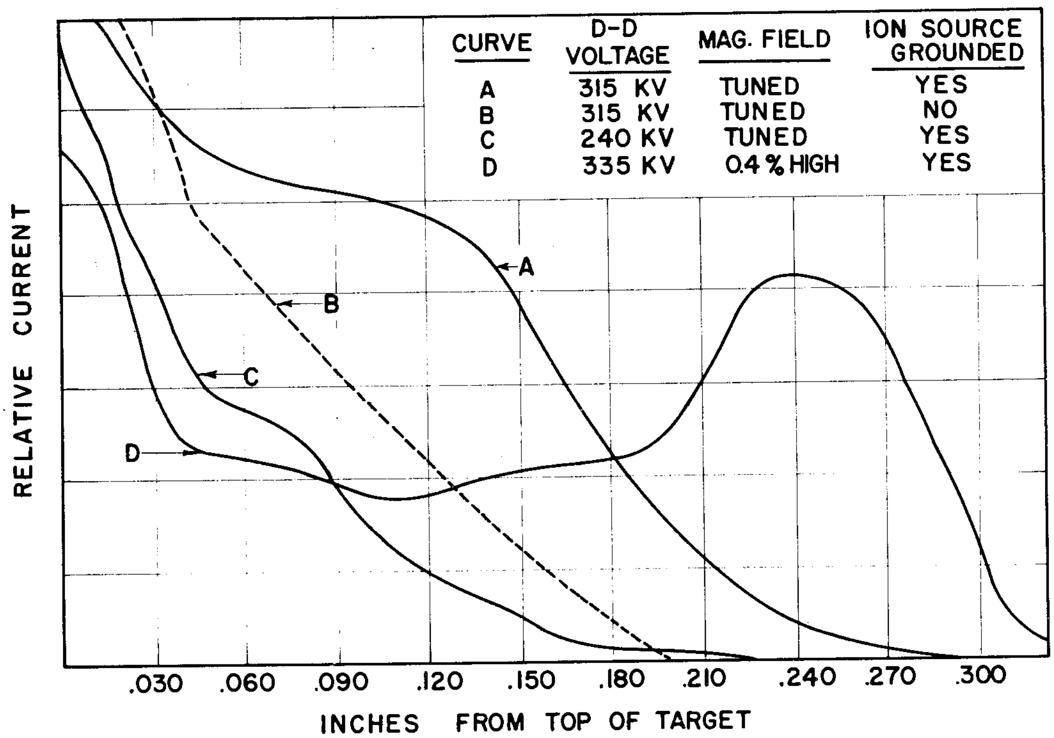
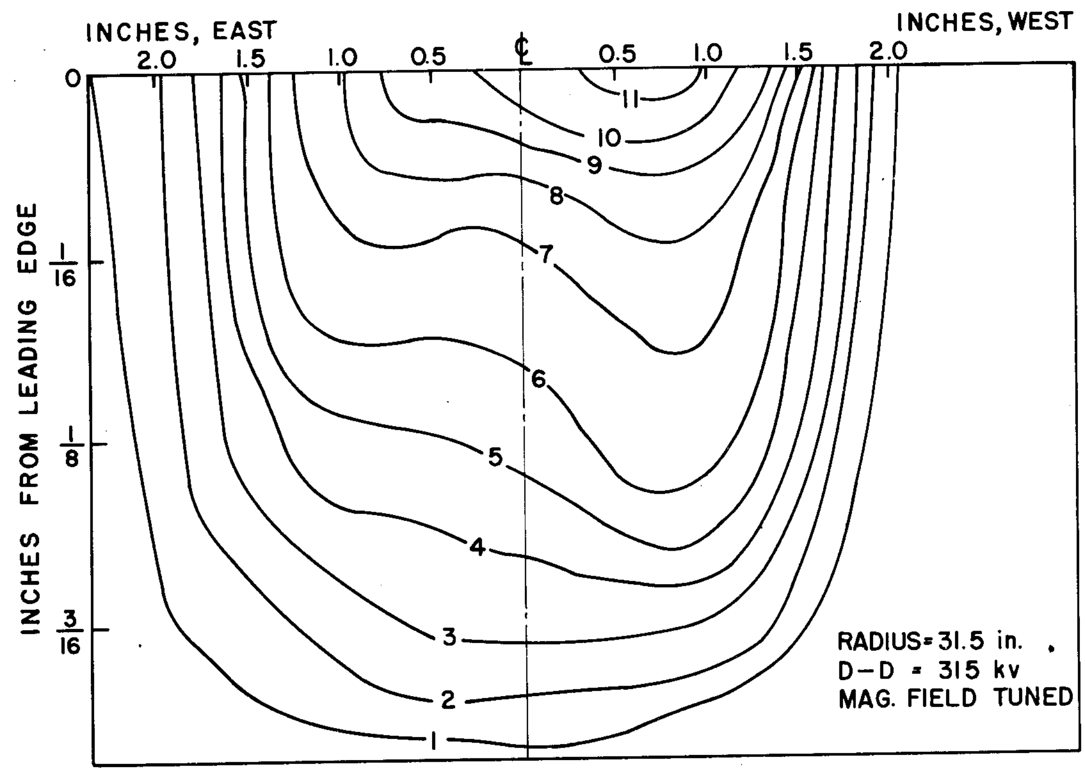


FIGURE 1. BEAM PATTERN TARGET



a. RADIAL DISTRIBUTIONS AT 30.5 INCHES



INTENSITY DISTRIBUTION ON TARGET

b. PATTERN ON TARGET, FOR A, ABOVE

FIGURE 2. INTENSITY DISTRIBUTION OF BEAM

neutron flux, normalized to a 1 ma proton beam on tantalum at 25 Mev, is tabulated below in units of permissible exposure (200 n/cm^2-sec for a 40-hour week).

	Units
Waist height, center of door	0.1
Waist height, edge of door	6
Waist height, 4 feet in any direction	0.2
On floor, center of door	0.5
10 feet above floor, center of door	60

Radiation Level in Cyclotron Pit

An experimental basis for estimating the ultimate level of radiation in the cyclotron pit was reported previously.* From subsequent experiments using more uniform and larger proton currents at higher energies it is evident that previous estimates of activities were too high by a factor of 1.5 to 2, except for the Fe 59 activity. For this activity the handbook value for manganese in SAE 1010 steel is too low; a recent spectrographic analysis of scrapings from the pole faces indicates a considerably higher manganese content, 0.6%. Also, since the dees are now lined with graphite sheets, it is expected that the Zn 65 activity will be eliminated. In the revised table below, the expected radiation level near the cyclotron after very long bombardment is normalized to 1 ma of 25 Mev protons on a tantalum target.

Half-Life	Source of Activity	mr/hr		
		Immediately	Next Day	3 Days After
12.8 hr	Slow neutron capturing in Cu	800	200	12
2.6 hr	Slow neutron capture in Mn in iron	1,200	1	--
45 days	Slow neutron capture in Fe	80	80	80

* Electromagnetic Research Division, ORNL, Progress Report for period ending March, 1951, p. 11, Y-767.

According to recent measurements the expected activity from Fe 59 at various places in the cyclotron pit, normalized as above, is:

Position	Expected Radiation Level (mr/hr)
Control desk	3
Balcony in front of oscillator	8
Cyclotron side of target changing shield wall	12
Back side of same	4
Far corners of pit	2
Near cyclotron	80
Between polefaces (dees and liner out)	800
Top of vacuum tank	6

Measurements made during a shutdown following a 1000 ma-hr bombardment indicate the presence of a long-lived activity 50% higher than this estimate, a very reasonable agreement considering assumptions used for the calculation.

Activity on Cyclotron Dees

Two sets of dees are now available for use in the cyclotron. While the second set has been in use the induced activity in the first set has been plotted and the decay followed. A prominent gamma activity due to 12.8 hour Cu 64 could result from Cu 65(p,pn) or Cu 63(n,γ). A strong long-lived ($\sim$200 days) activity is probably due to 250 day Zn 65 from Cu 65(p,n) and 270 day Ag 109(n,γ); these elements are present in the copper and in the solder used in the dees. The distribution of long-lived activities over the dees indicates high activity near the target and near a strut on the dee periphery, Figure 3 . There was evidence that this strut had intercepted high energy protons. It appears that a large part of the activity is neutron induced. Measurements of the beta-to-gamma ratio inside the dees indicated that activity produced by protons striking the copper of the dees was low.

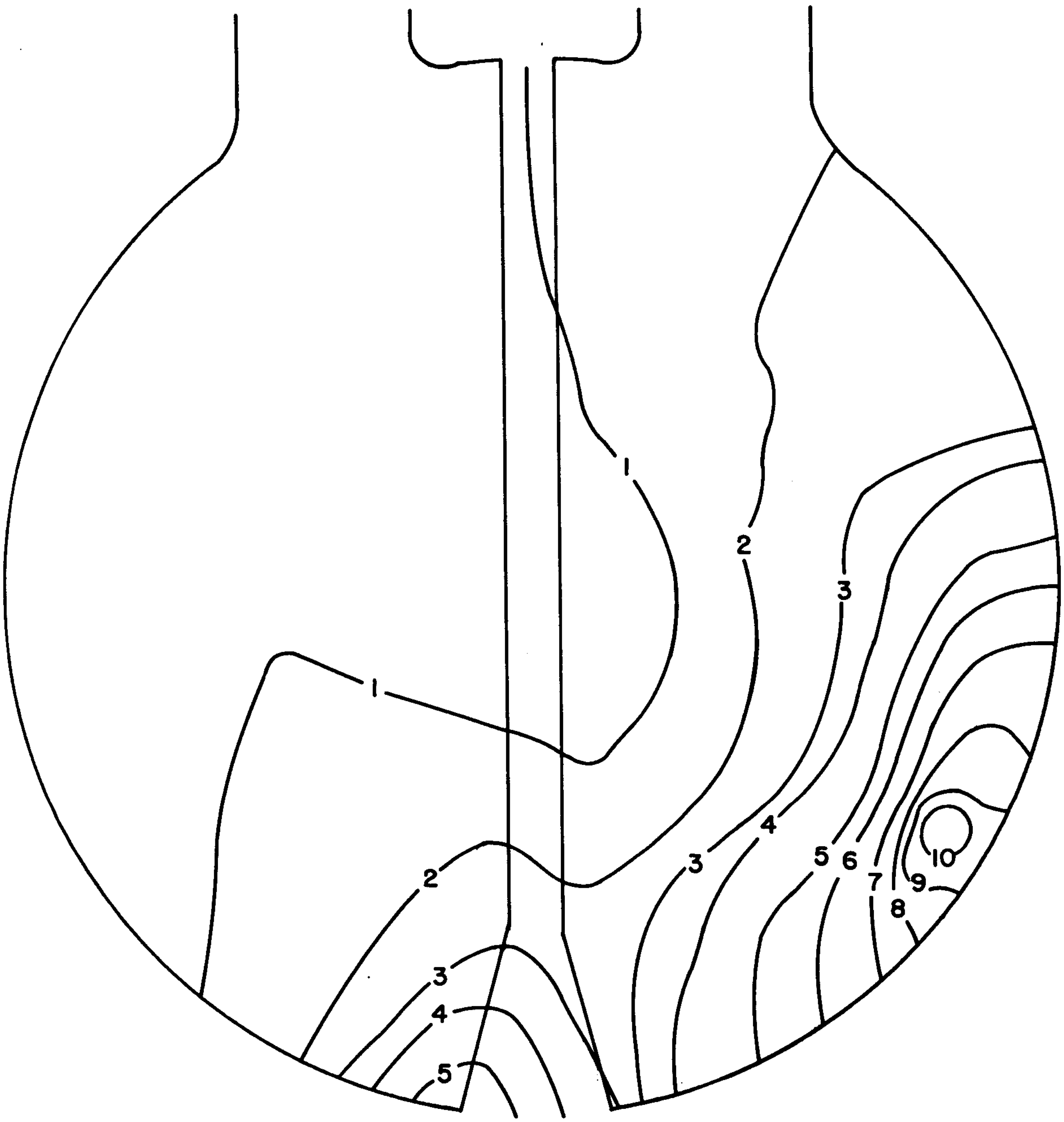


FIGURE 3. GAMMA RADIATION FROM DEE SURFACES

Activity on the dee constitutes a hazard to service mechanics and efforts have been made to reduce it. The short-lived activity is not serious since it readily decays to permissible levels. To eliminate the proton-induced activities the insides of the dees were completely covered with graphite sheets; since the neutron from protons on carbon is extremely low, this will also reduce the neutron-induced activity. In order to reduce the long-lived neutron-induced activity, future cyclotron dees will be fabricated from copper which is very low in undesired impurities, and silver solder will be avoided. Calculations have been made for the maximum amount of various impurities that can be allowed in order that saturation bombardment with 5 ma of protons on an average target will result in a neutron-induced activity radiation level of less than 500 mr/hr near the dees. In these calculations thermal neutron capture cross sections were used. Copper meeting these specifications, listed below, is commercially available.

Impurity	Half Life of Activity	Maximum Amount (ppm)
Ag	270 days	170
Au	2.7 days	30
As	1.0 days	1200
Sb	60 days	400
Se	127 days	600
Te	60 days	2,000
Fe	45 days	20,000
Zn	250 days	2,000
Co	5 years	6
Ta	117 days	20

Energy Measurements

Since the orbital center of the cyclotron beam does not coincide with the center of the magnetic field the radial position of the target from the center of the field cannot be used dependably as a measure of the energy of protons in the beam. The beam center is approximately 1.8" below and 2.5" south of the magnetic center. (The median plane of the magnetic field is vertical, with a north-south orientation.) As a result the beam strikes a strut in a dee structure when high energy is attempted. The energy of the beam at 30.5" from the magnetic center is somewhat less than 20 Mev, as measured both by photographic method and by the radioactivity induced in selected targets.

Photographic Method. Photographic film covered with a stepped aluminum absorber made from folded foil is wrapped in aluminum foil and exposed to the cyclotron beam. For this test the cyclotron beam is reduced to less than 0.1 μa by reducing ion source conditions and by considerable detuning of the magnetic field. Exposures of about 0.1 μa-sec were found suitable for a film having a Weston speed of 5. After the film is developed, the density for each absorber step is measured with a densitometer. From the plot of photographic densities the energy distribution of protons is calculated.

Measurements obtained by this method indicate that for a radius of 30.5" the proton distribution peaks at 18.7 Mev and has a width of 0.8 Mev at one-half height. Since the exposures are made under abnormal operating conditions of cyclotron operation the results are not readily reproducible. For this reason the photographic method has been largely abandoned in favor of a method using the excitation function of copper.

Excitation Function Method, Direct Target. Since the excitation function for Cu 63(p,n)Zn 63 $\rightarrow$ Cu 63 + β^+ (38 min) has recently been measured with high precision* the energy distribution of protons in the cyclotron is now being routinely determined by measuring the excitation function and comparing it with the published data. A stack of copper foils is bombarded through a

* Blaser, et. al, Helv Phys Acta 24, 1, 1951.

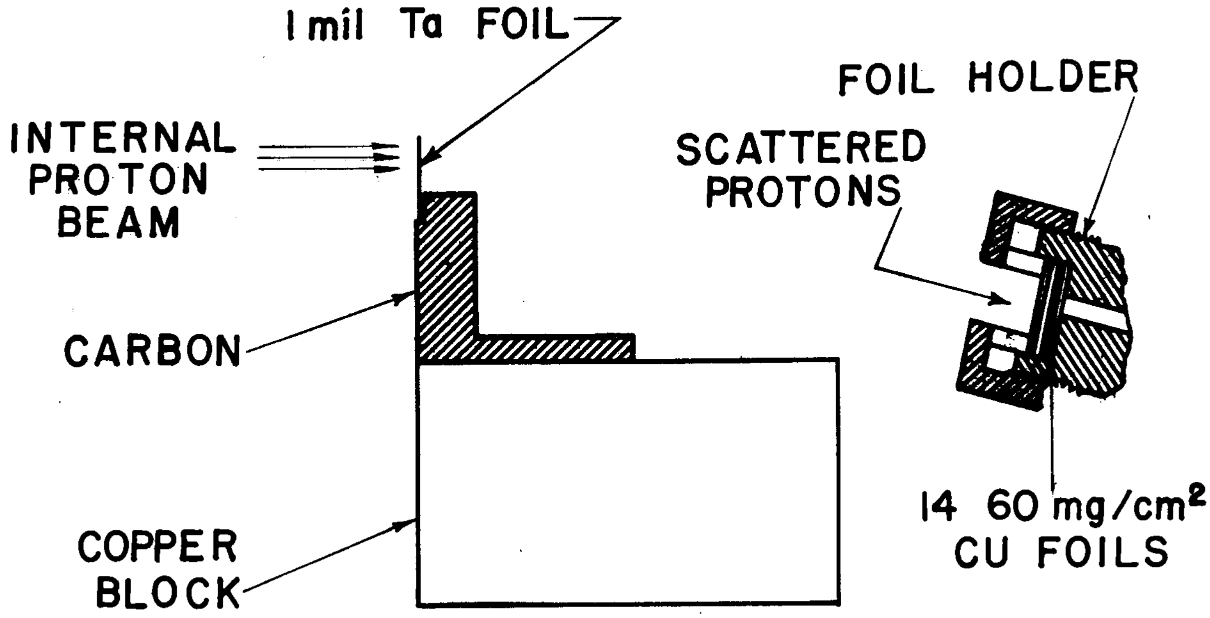


FIGURE 4a PROTON-SCATTERING TARGET

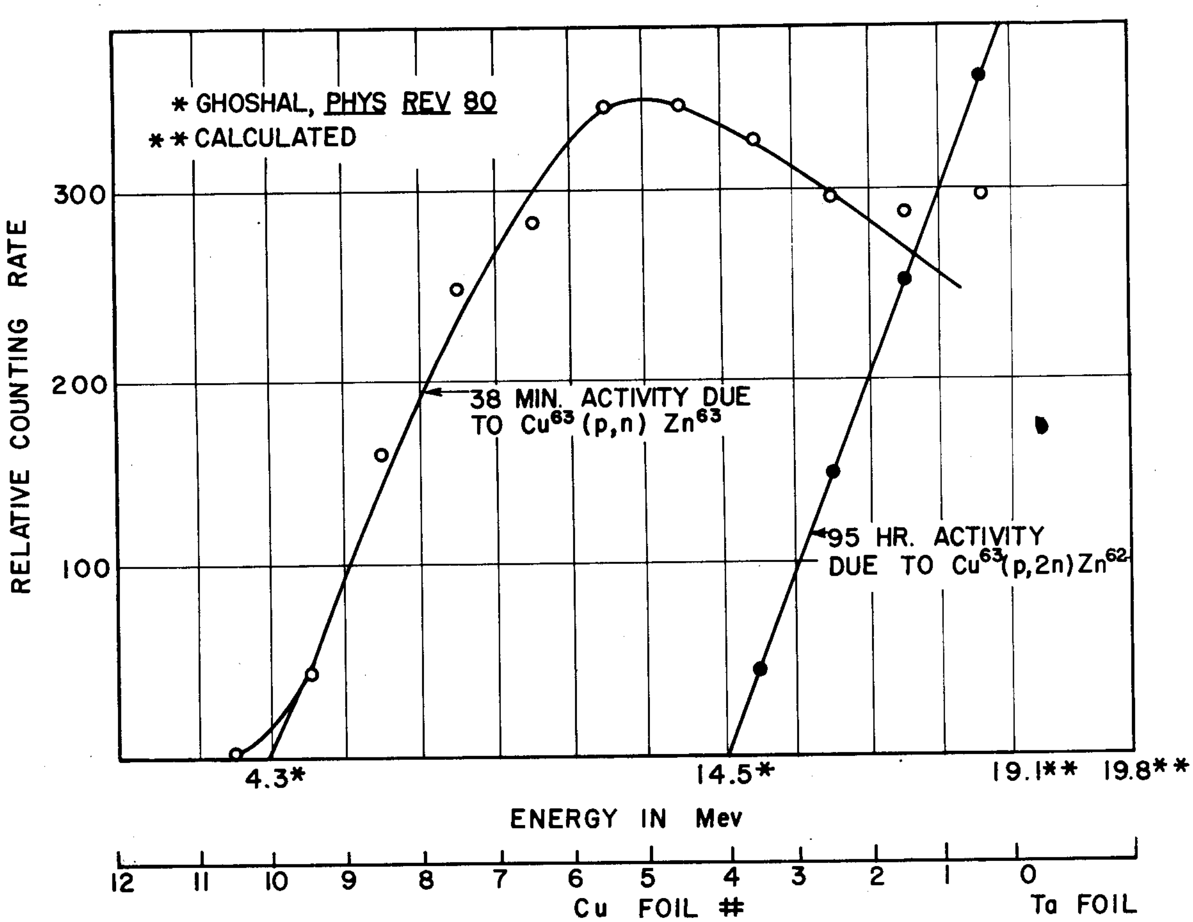


FIGURE 4b. ENERGY CALCULATION

tantalum window, Figure 4a, and the 38-min activity is determined as a function of depth in the stack. Line or block proton spectra which reproduce this distribution are calculated and from these the true continuous spectrum can be estimated, Figure 4b. This method has disadvantages in that ion source conditions must be adjusted to give low beam current and the edge of the window intercepts a portion of the beam. An advantage is that other targets may be bombarded at the same time.

Excitation Function Method, Scattering Target. The energy of the entire beam can be measured at normal operating conditions, but at reduced accuracy, by use of a target arrangement shown in Figure 5a. The activity induced in the copper foils by the scattered protons can be fitted to the published data on (p,n) and (p,2n) cross sections to determine the proton energy, Figure 5b.

NUCLEAR PHYSICS RESEARCH

Fission Asymmetry as a Function of Excitation Energy of the Compound Nucleus

A preliminary investigation of asymmetry of fragment distribution from proton-induced fission has been made as a function of proton energy in the region 11-18 Mev. Foils of normal uranium (11 mg/cm^2) separated by aluminum foils were bombarded at a fixed radius with the internal beam of the 86-inch cyclotron. The energy of the proton beam at the bombarding radius was found by measuring the excitation function of the (p,n) and (p,2n) reactions of Cu 63. The proton energy was deduced from the known cross sections of these reactions* and the range energy relation in copper. The 2.5 cm x 0.6 cm foils

* Ghoshal, S. N., Phys Rev 80, 939 (1950).

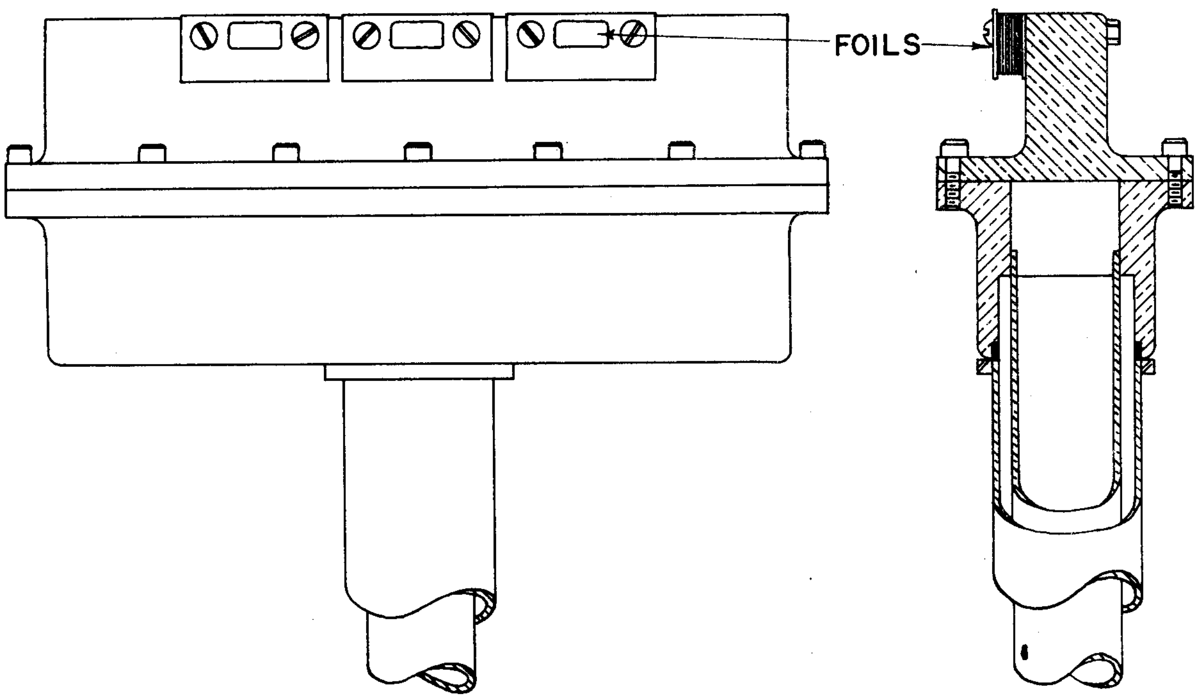


FIGURE 5a. STACKED FOIL TARGET

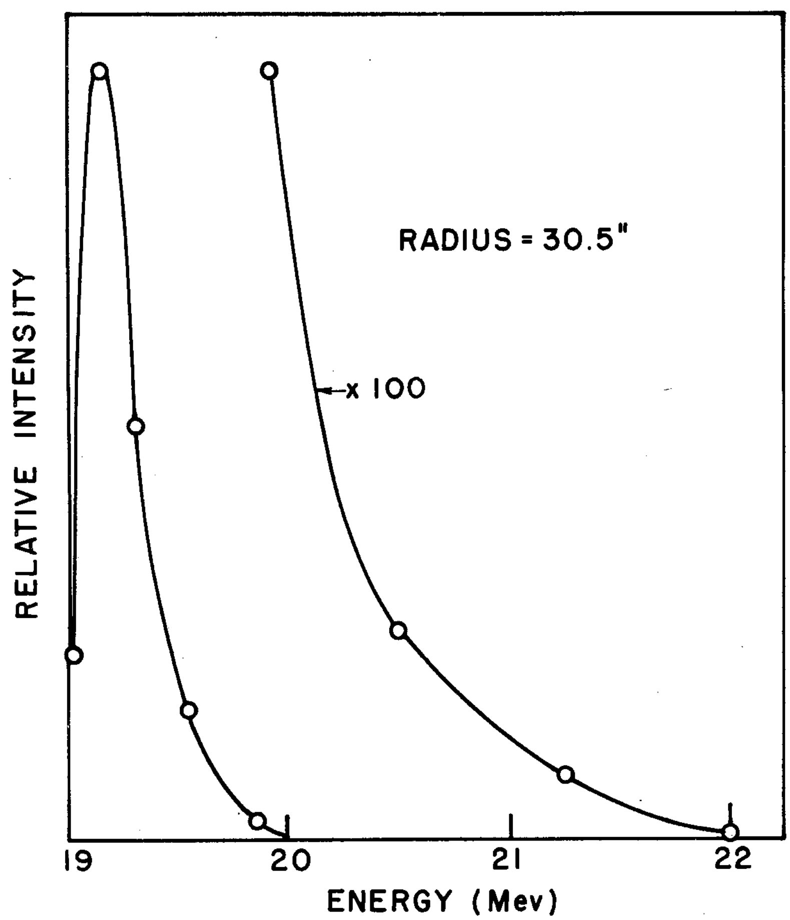


FIGURE 5b ENERGY DISTRIBUTION

of uranium and aluminum were supported behind a 0.011" window of a water-cooled aluminum probe, and the energy of the protons at each foil was calculated from the incident proton energy and the range energy curves in uranium and aluminum.

For each foil the fission yields of Mo 99 and Ag 111 relative to Ba 140 were determined by use of the radiochemical procedures.* Mo 99 and Ba 140 occur at the peaks of the thermal neutron fission mass distribution; Ag 111 occurs at the valley in this distribution. Relative yields were obtained in terms of known yields by performing the identical radiochemical analysis for thermal neutron fission in similar quantities of normal uranium. Neutron fission background was found in foils placed beyond the proton range.

The ratio, yield Mg 99/yield Ba 140, is almost constant as a function of proton energy and is about 60% higher than in the case of thermal neutron fission. The ratio of yield Ag 111/yield Ba 140, which is 0.0029 for thermal neutron fission,** varies rapidly with proton energy. This latter ratio (approximately the ratio of the minimum to maximum of the fission mass distribution curve) is plotted on semi-log paper against the reciprocal of the excitation energy of the compound nucleus, Figure 6. The particle binding energy is found from semi-empirical mass formulas.***

The same quantities for fission produced by other means are included for comparison. It appears that for the same compound nucleus undergoing fission, the ratio of the minimum to the maximum of the mass distribution curve varies

* Ghoshal, S. N., <u>Phys Rev</u> <u>80</u>, 939, 1950.
** Meinke, W. W., AECD-2738.
***<u>Rev Mod Phys</u> <u>18</u>, 513, 1946.

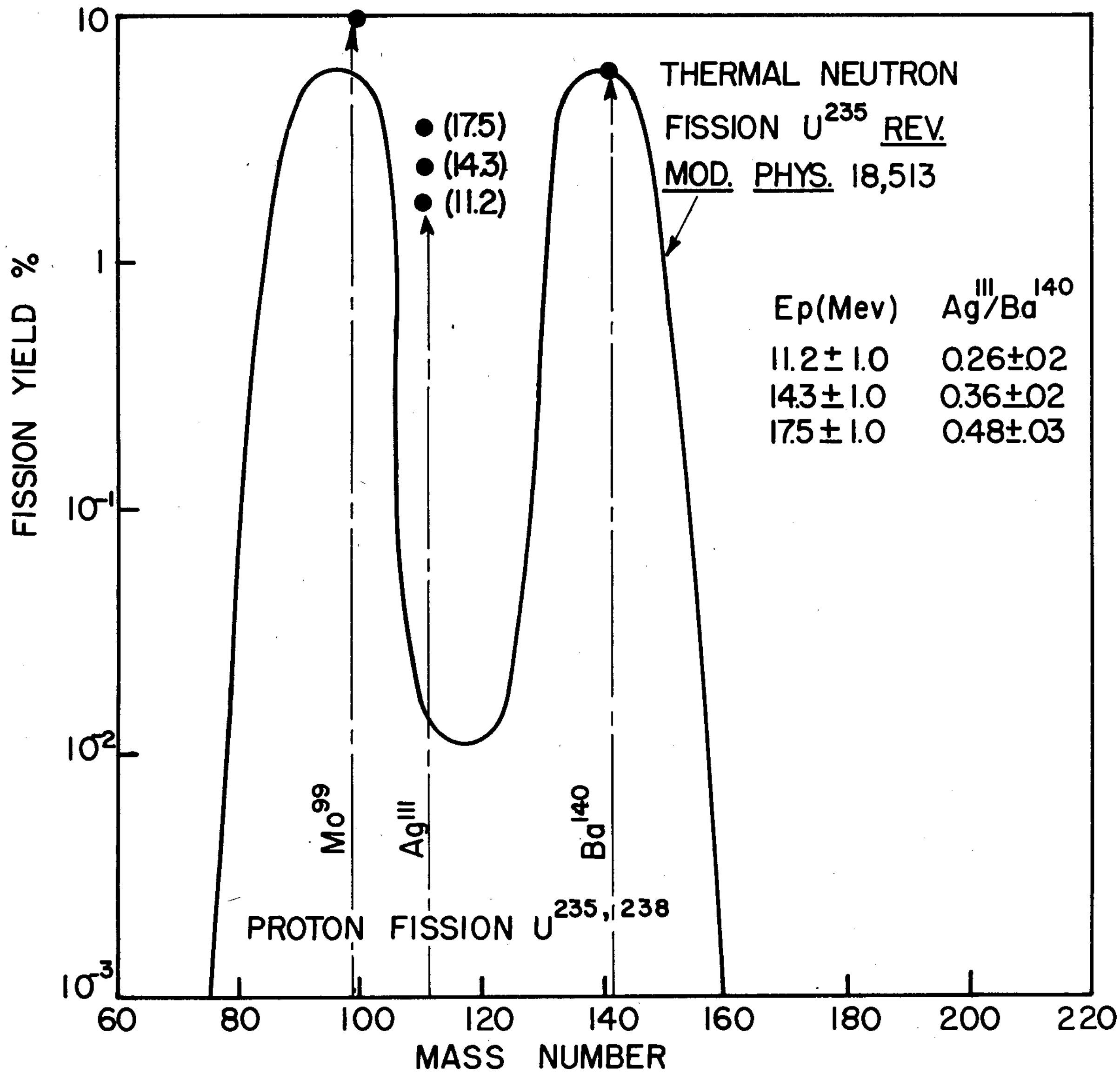


FIGURE 6. YIELD FROM PROTON-INDUCED FISSION

with compound nucleus excitation energy as $\sim ae^{-u/E}$, where a is $\sim$ 2.5 Mev and u $\sim$ 46 Mev in the case of U 236 fission. In the case fission of Np 236,239 (U 235,238 + p) u remains the same within the accuracy of the measurement and a is $\sim$ 3.3.

References for Figure 6 are given below:

1. Jones, Fowler, and Paehler, prepared for S. E. Section of APS, April 1952.
2. Rev Mod Phys 18, 513, 1946.
3. R. W. Spence, BNL L-C-9.
4. Newton, A. S., Phys Rev 75, 17, 1949.
5. Turkevich and Niday, Phys Rev 84, 52, 1951.
6. Engelkemeir, Seiler, Steinberg, Winsberg, Paper 218, National Nuclear Energy Series, The Fission Products, Div. IV, Vol. 9.
7. O'Connor and Seaborg, Phys Rev 74, 1189.

RADIATION DAMAGE

In continuation of the investigation of the effect of protons on Inconel fuel tubes containing uranium fluoride eutectic, sixteen fuel tubes have been irradiated with protons in the 86-inch cyclotron at $\sim$ 19 Mev. The power input during irradiation varied for individual bombardments from 150 watts/cc to a maximum of 415 watts/cc and the time of irradiation varied from one to eight hours.

Changes in the elastic limit of polycrystalline copper due to proton bombardment in the 86-inch cyclotron have been observed; the effects are compared with changes produced by neutron irradiation in a nuclear reactor.

TESTS ON INCONEL FUEL TUBES

Power Measurements

Two techniques were used in measuring the total amount of power delivered to the fuel-element target by the proton beam. Determination of the specific energy dissipation was then made by combining results of measurement of the bombarded volume of uranium eutectic fuel in the target with measurement of proton beam current.

Because bombardment of the fuel pin targets heats the fuel to above the design temperature, it is necessary to cool the fuel pins by brazing them to a copper tube through which cool water is continuously circulated. At the temperatures of this experiment (approximately 800°C), almost all of the power of the proton beam appears in the cooling water stream. Therefore, by measuring the entrance and exit water temperatures and the water flow rate it is possible to determine the power delivered to the target. However, because the apparatus used for cooling the target is extensive and not well isolated thermally, it is desirable to calibrate the actual system. For this calibration, measured quantities of heat were introduced into the thermally insulated water-cooled target mount by means of electric heating elements. From the known power input, measured water flow rates, and temperature change a correlation between the factors was obtained. This was then used for the determination of most of the power input measurements reported in this series.

A second beam current measurement, more commonly used, involves the insertion of a microammeter between the target and ground potentials. For routine low current bombardments this method is not useful because the target is customarily connected to ground through a thermocouple circuit and because both secondary and thermal electron emission from the target give the appearance of increased proton current. Also, oscillating electrons, constrained to cycloidal paths by the electric and magnetic field, may migrate to the target and further reduce the proton signal. Consequently, small values of proton current cannot be measured accurately.

In order to compare the power as measured by calorimetric methods with measurements by the more direct electrical method, a special target housing was devised. This housing consists of an outer graphite box at ground potential, Figure 7. A diagonal slot parallel to the target at front and rear permitted passage of the proton beam to the target. Two inner graphite sheets are mounted at right angles to the axis of the target and are connected electrically to the target and insulated from ground. During proton bombardment the outer housing shields the components at target potential from oscillating electrons; the inner graphite members serve to collect the secondary and thermal electrons emitted at the target; and the net electrical effect of the proton bombardment is read on the target current meter.

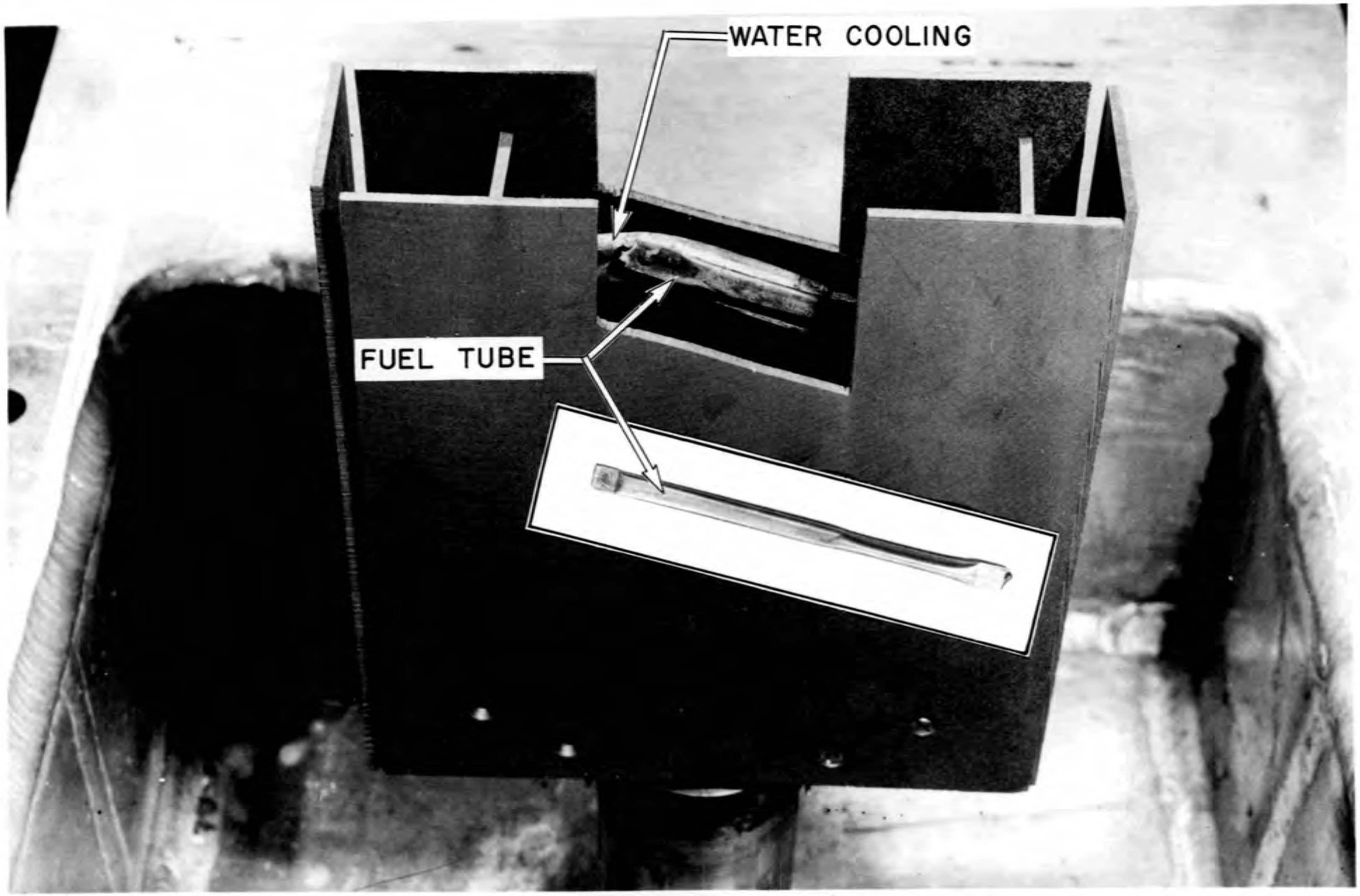


FIGURE 7 RADIATION DAMAGE TARGET

A stainless steel plate mounted on the water-cooled backing in this housing was then bombarded with 19 Mev protons. Energy inputs to the target were determined calorimetrically and compared with those read on the target meter. Agreement over a wide range of proton beam currents was always within eight per cent and was considered adequate for these experiments.

Limitations of Power Input

The power input achieved in the fuel tube irradiation experiment was a factor of ten less than the proposed goal for specific power dissipation. Although the techniques described permit a considerably closer approach to the desired 4000-5000 watts/cc, the effective limitation on the power input is the thermal conductivity of the eutectic fuel itself. This low conductivity recently reported as 0.53 Btu/ft^2-hr-°F/ft, indicates that the fuel is about as good a thermal insulator as rock wool. Calculations based on this value show that under the conditions of the test only about 350 watts/cc could be conducted through the fuel. Within the accuracy of the calculations and measurements the results of this series of bombardments seem to be consistent with this conclusion; the maximum power density dissipated in any target was 415 watts/cc. Because of the limitations inherent in the system of water cooling used it is probably not possible to enhance the effective thermal conductivity by introduction of convective currents within the target. The temperature of the water stream, and hence that of the surface of the fuel pin, is approximately 15°C, and since the eutectic is solid at temperatures below 500°C, it is probable that a crystalline layer of eutectic at the rear surface of the target prevented formation of an effective convection loop.

Chemical Analysis

The nature of the chemical analysis of the eutectic fuel was such as to yield two types of information. The uranium and fluoride content of the fuel mixture in each specimen was assayed in an effort to measure the extent of radiation-induced chemical decomposition of the fuel fluorides. A decrease in fluorine content in the eutectic serves to indicate the extent to which such decomposition is followed by recombination with the Inconel wall. The second analysis of the fuel mixture was made to indicate quantitative information about the nature of the decomposition of the metal tube wall. Trace quantity analysis for the elemental Inconel components was made to determine rate of solubility of each of these elements independently under irradiation.

Results of the quantitative uranium and fluorine determinations show that all values--irradiated samples, heat-treated samples, and original batch analysis--lay within ±1% of the mean. Being well within the accuracy claimed for this analysis, the results were interprěted as indicating no radiation induced effect on the relative amount of uranium and fluorine in the eutectic hence, no decomposition of the eutectic over the limited range of irradiation power inputs of these experiments.

Results of the quantitative analysis for elemental Inconel components showed no significant difference between heat treated samples and proton irradiated samples. No effect on the amount of iron in the eutectic was found in either case. Chromium, which seemed to be selectively soluble in molten eutectic, showed no additional effect due to proton irradiation. Analyses do show a decrease in the amount of nickel in the eutectic as a result of heat treatment and proton irradiation.

Metallurgical Analyses

Several of the earliest fuel pin targets gave evidence of grain growth, a property which occurred only once in a control sample. To investigate the possibility that the effect was a result of irradiation a series of experiments was performed. In the first experiment samples of the Inconel tubing in "as received" condition were heated for one hour; individual samples were heated at 100°C temperature intervals, a total of eight covering the temperature range from 500°C to 1200°C. Metallurgical examination showed that grain growth was produced under these conditions at temperatures above 900°C. A second series of samples made from the same stock was reshaped to duplicate the geometry and cold working of the cyclotron targets. Results of these tests, again in the range 500°C to 1200°C, were the same as for the earlier series. The grain growth above 900°C in these tubes duplicated the effect of the cyclotron series, and eliminated the possibility of its being uniquely radiation-induced. Grain growth in the early targets is attributed to inhomogeneities in the cyclotron beam, with consequent poor temperature regulation, local overheating, and grain growth.

To date, none of the metallurgical properties examined over the limited range of irradiation power inputs of these experiments show any effects ascribable to irradiation.

COMPARISON OF NEUTRON AND PROTON EFFECTS ON COPPER

While the 86-inch cyclotron has been used for empirical studies of radiation damage no neutron-proton correlation has been made to date. The work of T. H. Blewitt* has shown that a suitable attack on this problem would be an examination of the plastic properties of copper. In particular,

* Physics of Solids Institute, ORNL, Quarterly Progress Report for period ending January 1951, ORNL-1025, p. 22.

his work has shown that the critical shear stress in single copper crystals is sensitive to reactor radiations. In order to pursue these ideas in a convenient, yet rapid, fashion polycrystalline copper is used instead of crystalline copper. The polycrystalline property analogous to critical shear in crystalline material is the elastic limit; this has also been shown by Blewitt to be sensitive to radiation.

After some preliminary bombardments an existing type of cyclotron target head (Type A-1) was found suitable. The target is mounted inside the water channel of this target head and is held by springs against a 0.010" aluminum window through which the proton beam enters. This arrangement provides efficient target cooling but there is no provision for thermocouple leads to the target so the current distribution on the target must be estimated from previous measurements for this type of target.

For the first test, two identical polycrystalline copper tensile specimens, 0.016" thick, were annealed at 1000°C. One was irradiated for an estimated 2.5 μa-hr/cm^2 while the other was retained as a control. The elastic limit was 4.6 kg/mm^2 for the irradiated sample, as compared with 1.1 kg/mm^2 for the unirradiated sample. The two specimens were again annealed at 1000°C and then one specimen was irradiated for an estimated 5 μa-hr/cm^2 while the second was mounted behind the first with an aluminum absorber in between, thick enough to stop the protons. Unfortunately, the control specimen was bent by the flow of cooling water and the observed elastic limit increased from 1.1 kg/mm^2 to 1.8 kg/mm^2. The value for the irradiated sample was 5.4 kg/mm^2.

Irradiations of 1×10^{18} nvt in the X-pile were observed by Blewitt* to increase the elastic limit of polycrystalline specimens by a factor of 2.5. His data on pure crystals indicates a roughly linear increase in the factor to 1×10^{18} nvt, where saturation sets in. Although more measurements should be taken, estimates based upon cyclotron data are that saturation sets in at about 2.5 μa-hr/cm^2. A reasonable control value would be an average of unmounted and mounted control samples of 1.5 kg/mm^2. Then a logical datum for comparison with pile data is 2.5 μa-hr/cm^2, causing a change by a factor of 3.1 (with about 50% error) in the elastic limit. Thus, experimentally, 1×10^{18} nvt $\cong$ 2 μa-hr/cm^2.

Seitz's calculations** make use of a critical point where elastic-energy transfers equal ionization and excitation transfers. For copper, this occurs for 800 kev recoils or 26 Mev neutrons, too high for pile irradiations. Preliminary results of a recalculation, using a 1/E spectrum and certain averaging processes, indicate 1×10^{18} nvt = 0.5 μa-hr/cm^2, in order-of-magnitude agreement with the above result.

A further result of these calculations is that the high energy neutrons of the pile contribute about as much, per unit energy range, to atomic displacements as the low energy epithermal neutrons. This follows since the energy transferred by the neutron (all in elastic collisions) varies as E and the flux as 1/E, the collision cross section remains comparatively constant, and the fraction of energy lost by the recoils to displacements is also comparatively constant. Neutrons from a (p,n) reaction should then be more effective, per unit flux, in producing displacements than pile neutrons. An

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* Private communication.

** Seitz, F., TID-65, p. 142.

irradiation using (p,n) neutrons led to an elastic limit change by a factor of 1.7 for an estimated 10^{16} nvt, showing an apparent advantage of 60 times (again per unit flux) over a pile bombardment.

The conclusion that comparatively modest beam currents produce radiation damage effects (which depend on displacements) many times faster than pile irradiations is substantiated. If one finds it convenient to use neutron irradiations, fast neutrons from beryllium targets may be used as effectively as neutrons from the LITR.

ELECTRO-NUCLEAR MACHINES

Components of the 63-inch cyclotron have been fabricated and the machine is now being assembled and tested. On the 22-inch test cyclotron, 38% of a 1000 μa internal beam has been deflected; an examination of the fine structure of the proton beam along the radius indicates distinct proton orbits.

THE 63-INCH CYCLOTRON

Fabrication of the 63-inch cyclotron components has been completed, and the machine is now being assembled and tested. The cyclotron liner and the liner faceplate are ready for installation and the dees with their 17-foot dee stems are ready to mount on the faceplate, Figure 8.

Nuclear Reactions

Since it is probable that the 63-inch cyclotron will be operated for some time with an internal beam, a solid nitrogen target has been developed. Tantalum nitride is produced by heating tantalum foil to 1100°C and then introducing nitrogen gas under pressure. The resulting brittle tantalum nitride foil can be broken into small plates suitable for cyclotron targets. Such targets can be clamped to a water-cooled probe and bombarded with the undeflected nitrogen beam. The 10-minute activity of N 13 can be detected after quick removal of the probe and target through the vacuum lock. In this way an estimate can be made of the cross section of the reaction N 14 + N 14 → N 13 + N 15. A proportional counter has been tested with a 5×10^{-5} inch nickel foil over a 1/8" opening. Such a foil has proved to be tight and the counter operates satisfactorily.

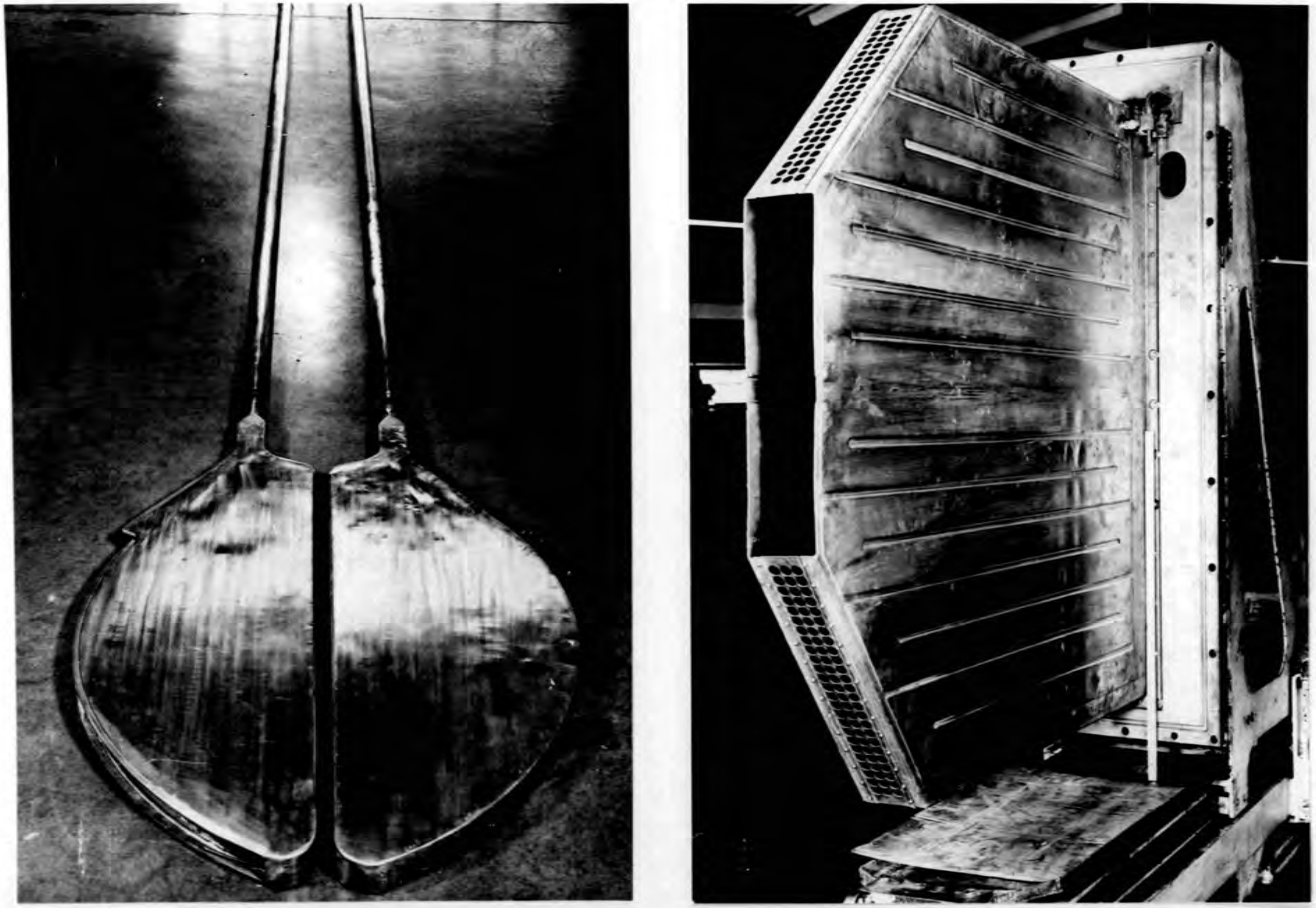

FIGURE 8 63" CYCLOTRON DEES AND LINER

A pulse height analyzing circuit consisting of a single channel pulse height analyzer, amplifier, preamplifier, scalers, and power supplies has been assembled. This apparatus was tested out on a beta-ray spectrometer with an anthracene crystal and a photo-multiplier tube. The beta spectrum of Cs 137 has been measured and the conversion line at 0.665 Mev observed.

Field Operations

Three windows have been provided in the vacuum tank for the 63-inch cyclotron. These windows will be used for observing such things as ion source conditions and rf arcing inside the tank; they are placed in such a position that probes can be inserted through them for measuring the position of the beam. The vacuum manifold has also been installed and two 20" diffusion pumps connected to the system. The pumps have a combined pumping speed of 13,500 liters/sec. A vacuum of 0.004 microns has been obtained in the tank, with a rate of rise of 0.0002 microns/sec. All the components of the cyclotron, such as dees and dee stems, were individually vacuum tested upon delivery from the shop in order to minimize leak troubles after assemblies have been completed.

The electrical connections for the operation of the cyclotron including high voltage, control, and interlock circuits, are now in place and the control console has been wired, Figure 9. The central section, shown at the left has two galvanometer scales, one for measuring the beam current and the other for monitoring the magnetic field. In addition there are magnetic field controls, ion source controls, and vacuum gauges. The section to the far left, not shown, will contain experimental equipment such as scalers, amplifiers, beam integrator, and a 10-channel pulse height analyzer. The three panels to the right of the central panel contain, in order: controls

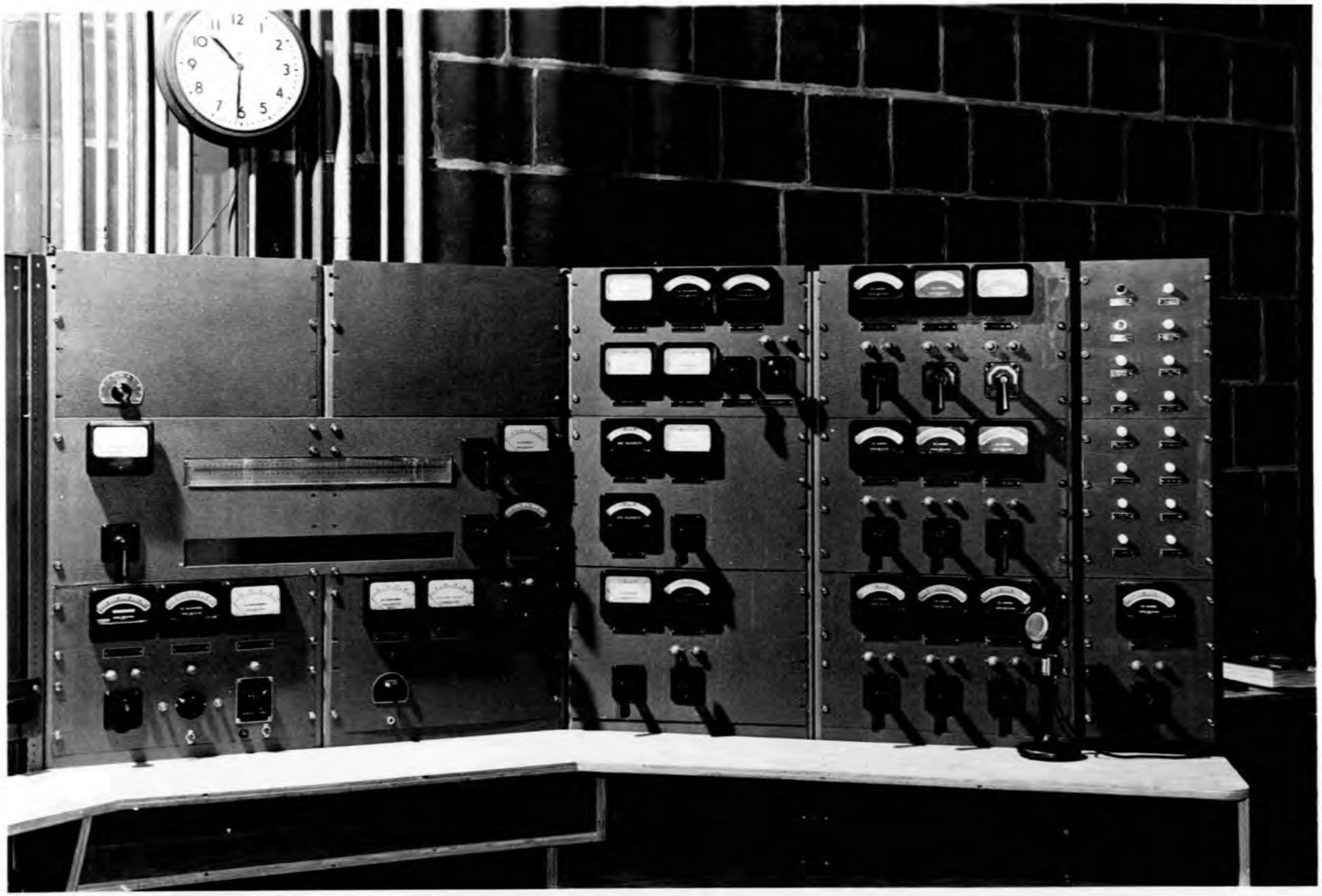

FIGURE 9 CONTROL CONSOLE, 63-INCH CYCLOTRON

for the rf system, dee voltage, dee bias, etc.; controls for the power supplies for the oscillator plate; and, to the far right, the interlock and indicator lights.

The Rf System

The construction, assembly, and wiring of the oscillator and coupling circuits has been completed. Figure 10 shows the oscillator cabinet with the top and access side doors removed. The ground plane separating the filament and plate circuits, typical of the grounded grid oscillator, is clearly shown. Filament bifilar rf chokes and filament blocking condensers are located in the upper compartment, while the plate neutralizing coil, by-passing condensers, and insulating Lapp coils are shown below. Plate-cooling water is fed through the Lapp coil and the plate neutralizing coil. This is more clearly shown in Figure 11a, along with the details of the tube mounting and the tube cooling air duct. The blower supplying air to the tube seals is mounted on the right side of the oscillator cabinet. The filament transformer is just above the blower and the filament coupling circuit is shown in Figure 10b. The plate coupling circuit is similar; both coupling circuits, as well as the oscillator, are mounted on casters to facilitate cyclotron assembly.

An initial test has been made on the oscillator tube air, water, and filament circuits. The test was satisfactory with respect to the cooling facilities but some difficulty, due to vibration, has been encountered in the case of the filament transformer. Work is now in progress to eliminate this source of noise as well as to reduce the vibration of the tube components. Tests will continue, as far as facilities will permit, until the dee resonant system has been assembled and is ready for rf tests.

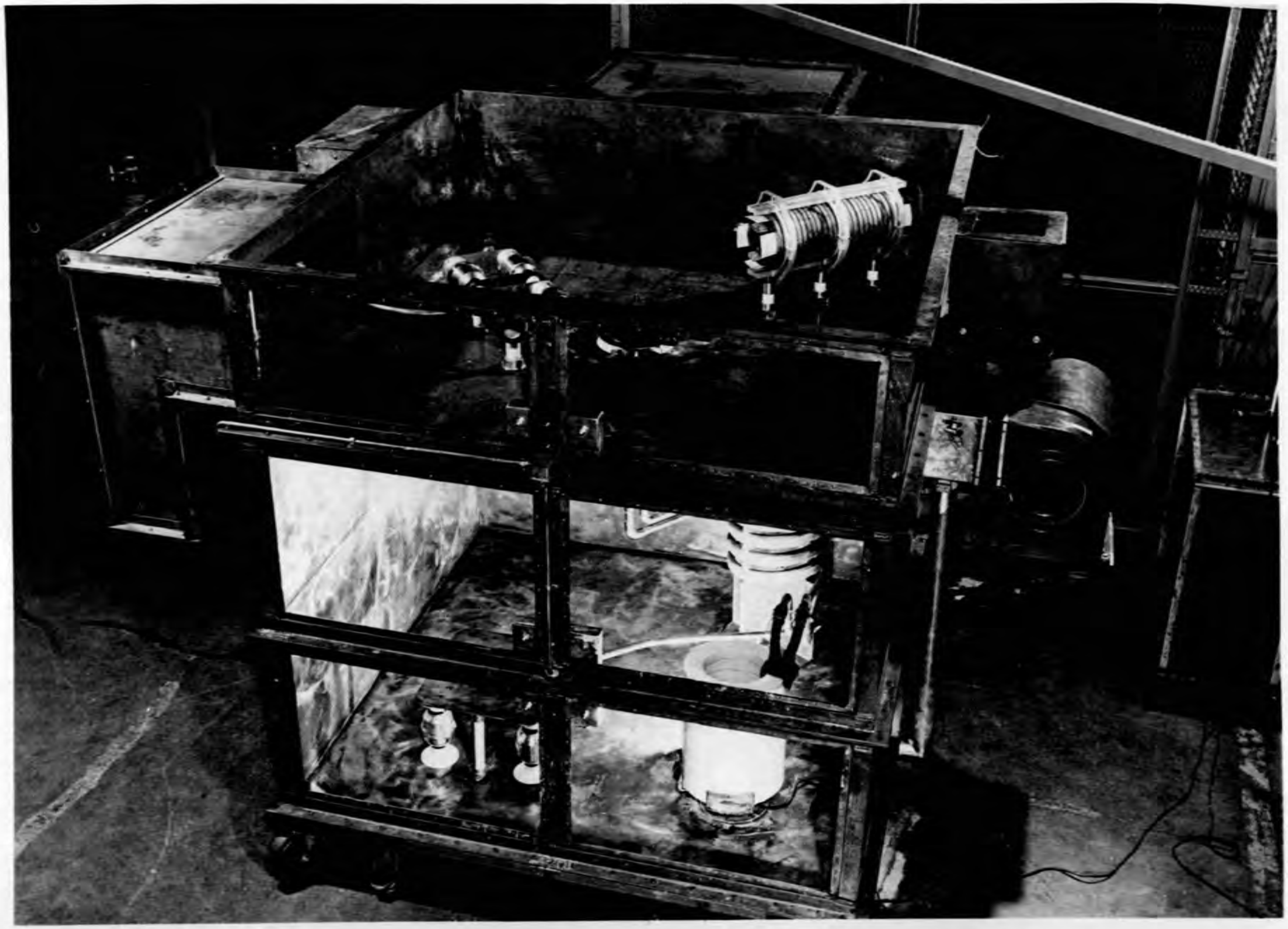

FIGURE 10 OSCILLATOR, 63-INCH CYCLOTRON

FIGURE 11a. PLATE SECTION

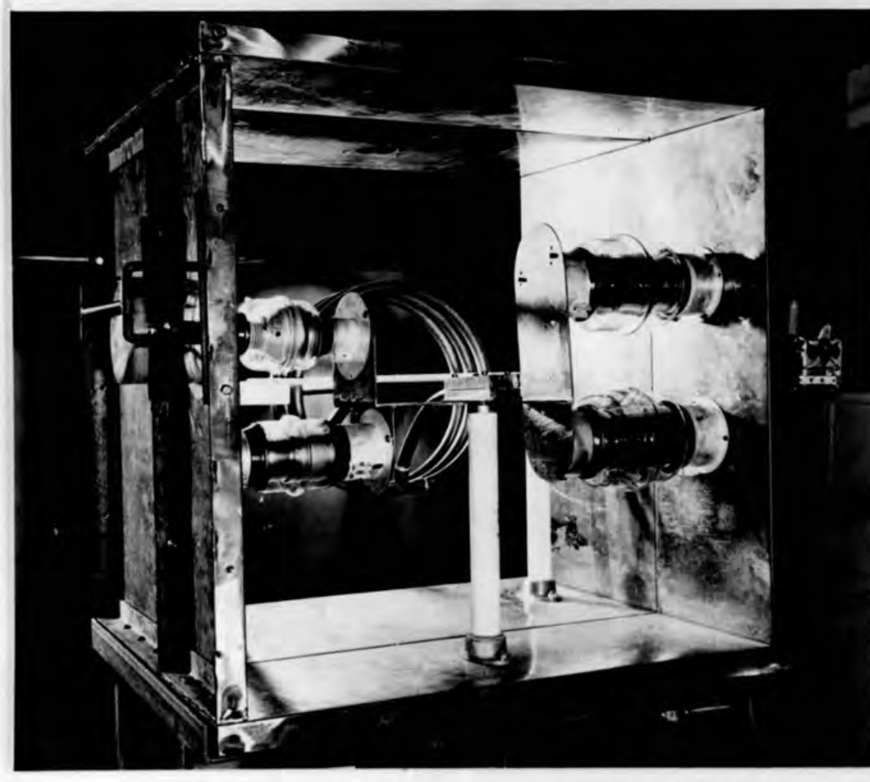

FIGURE 11b. FILAMENT COUPLING

THE 22-INCH TEST CYCLOTRON

An examination of the proton beam fine structure, radius-wise, has been made to determine the orbital pattern of the beam. This information will be used in the development of a grid designed for electrical focusing. A 0.020" tantalum wire is used as a movable probe to scan the beam radially from 1.2 to 11.5 inches. The data show individual orbital positions from the first orbit up to the twelfth, the upper limit being determined by the potential on the dees. With high dee voltage maximum currents have been observed to be separated by 5/8" for the inner orbits. The minimum current between orbits has never been observed to be less than one-third the maximum.

A typical fine structure of the beam is shown in Figure 12. In determinining the probe position for each maximum and each minimum the 0.020" probe was positioned manually. There is a slight increase in probe current with increasing radius because of increased thermal emission of electrons from the probe as it was bombarded by protons of higher energy.

A preliminary test of a dc injector-type ion source of the 22-inch cyclotron has indicated the need for modification of the accelerator electrode design. A very effective adjustable beam deflector has been developed; under the best conditions, 38% of a 1000 μa internal beam has been deflected.

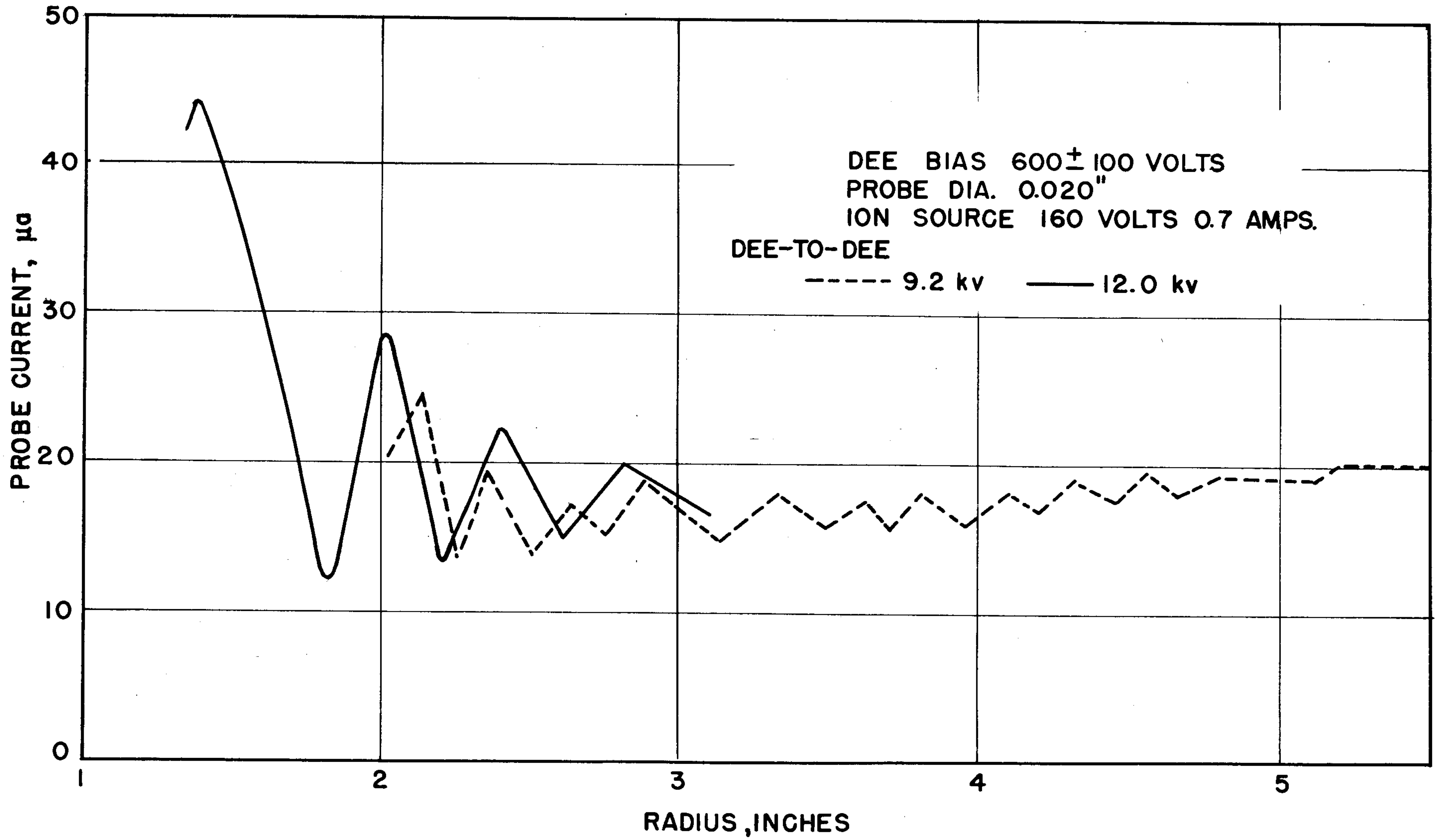

FIGURE 12 BEAM FINE STRUCTURE

SPECIAL SEPARATIONS

Several grams of very pure uranium 235 have been prepared and are now available for research purposes. An earlier separation of bismuth 210 has resulted in new determinations of the properties of a Bi 210 isomer. Final chemical recovery efficiency for the first stage of the U 236 separation program has been determined at 97% of throughput. Facilities are being prepared for laboratory-scale separation of plutonium isotopes. Eight shipments of enriched uranium isotopes have been supplied.

ENRICHMENT OF URANIUM 235

A quantity of U 235 low in U 234, produced as a by-product of U 236 enrichment, was reprocessed electromagnetically to deplete still further the U 234 content. Because of its high alpha activity the presence of U 234 in U 235 is particularly objectionable when specific activity and fine structure are being determined. In the purest U 235 sample available alpha radiation from U 234 was about 35% of the total activity.

In the reprocessing of U 235 the alpha activity due to U 234 has been reduced to approximately 3%. The mass fractions 234, 235, 236 and 238 were collected in the multiple-pocket collector developed for separating U 236. Estimated composition of uranium in the U 235 pocket is as follows:

Isotope	Isotopic Composition (%) Feed	Isotopic Composition (%) U 235 Product	Relative Alpha-Activity in Product (%)
U 235	99.81	99.994	97
U 236	0.06	0.004	0.1
U 238	0.10	0.001	0
U 234	0.03	0.001	3

Approximately 7.5 gms of U 235 was obtained from 79 gms of feed. This highly purified U 235 will be made available for use on Atomic Energy Commission projects as soon as it has been chemically purified, converted to the oxide, and analyzed.

BISMUTH ISOTOPIC ENRICHMENT

Neutron-irradiated bismuth was supplied by the University of California Radiation Laboratory for the enrichment of the alpha-active Bi 210 isomer which gave a count of $\sim$2.1 c/m per mg of bismuth. Late in 1950* the bismuth was electromagnetically separated and the mass fractions 208, 209, and 210 were collected. Upon further analysis the specific alpha activity of the bismuth in each mass fraction has been determined to be as follows:

Mass Fraction	Sample	c/m-mg Bi	Enhancement	
210	1	64.2	210:209	31
	2	71.3	210:209	34
209	1	Not detectable	209:210	15
	2	0.3	209:210	7
208	1	0.6	208:209	15
	2	0.9	208:210	50

With this enriched Bi 210, Levy and Perlman** were able to verify the existence of the long-lived alpha-active isomer of Bi 210 and to re-evaluate the decay scheme.

* Electromagnetic Research Division, ORNL, Quarterly Progress Report for period ending December 1950, p. 42, Y-722.

**Chemistry Division, UCRL, Quarterly Progress Report for period ending August 1951, p. 14, UCRL-1507.

SECOND STAGE U 236 SEPARATION - CHEMICAL RECYCLING RESULTS

The first stage of U 236 separation was followed by an intensive effort to recover holdup uranium from electromagnetic equipment, mainly because of the prospect of contaminating the second stage feed bank. From U 236 stocks produced in the initial stage, 26 grams of uranium at 32% U 236 were allocated to further enrichment. Prolonged recycling of the starting feed had changed the isotopic composition; for six cycles uranium cross-over into the second stage totaled 1.1 gm U 235 and 1.1 gm U 238. Collector efficiencies for uranium isotopes ranged from 8% for U 235 to 0.2% for U 238 for the two-pocket collector. The combined effect of non-uniform removal of isotopes as product and of gain in uranium-bearing material from previous separations is illustrated in the following table:

Isotope	Feed Concentration (%)		Product Concentration (%)	
	Start	End	Start	End
U 236	32.0	28.9	95.7	95.2
U 235	62.5	57.4	4.0	4.0
U 238	4.7	12.5	0.2	0.7
U 234	0.8	1.2	0.1	0.1

Between separation runs, about four-fifths of unresolved uranium was recovered for re-separation. On completion of the enrichment phase of the program, the unseparated residue totaled 22 grams of metal at 25% U 236. Terminal recovery efficiency was therefore 97% of the U 236 throughput.

FACILITIES FOR PROCESSING PLUTONIUM

Installation of facilities for the separation of plutonium isotopes on a laboratory scale is nearing completion in Building 9204-3. These facilities include a processing laboratory, a calutron wash area, and designated calutron equipment.

The CO_2 fire protection system, permanent air samplers, and the remaining gloved boxes have now been installed in the processing laboratory. A gloved enclosure for the evaporation equipment is being installed. Process equipment for multi-stage batch extraction of concentrated machine wash was completed and components for an ion exchange bed fabricated. The possible application of ion exchange for direct recovery of plutonium from calutron machine wash is being investigated; if successful, this will result in a considerable simplification of recycle chemistry.

The enclosure for the calutron wash area with its ventilation, services, and process piping was completed. The construction of gloved boxes and an acid spray system for washing calutrons is well advanced.

A calutron vacuum tank is being adapted for plutonium operation. The vacuum system of this tank was isolated from other vacuum equipment and filters installed for cleaning exhaust gases. To provide for safely removing a contaminated calutron from the tank, alterations have been made at the tank front to permit attachment of a vinylite bag which will cover calutron units as they are removed. A baffle was installed in the tank extension to minimize contamination of the outside of the calutron liner. Preparations are being made for subjecting the calutron to an atmosphere of chlorine at the end of each run, before exposure to air, in order to minimize the formation of insoluble oxides which would impede chemical recovery of plutonium.

Following the completion of the facilities extensive testing will be required to insure that the equipment functions adequately and that the gloved boxes and proposed transfer methods will properly contain the high alpha activity. Test runs with alpha-active U 234 and Th 230 are planned. In addition to their value as preliminary tests for plutonium operation these runs will yield useful quantities of concentrated isotopes.

SHIPMENTS OF ENRICHED URANIUM ISOTOPES

Distribution of electromagnetically separated uranium isotopes to Atomic Energy Commission laboratories during October, November, and December is tabulated below:

Oak Ridge National Laboratory	10 mg U	95.51% U 236
Argonne National Laboratory	10 mg U	95.31% U 236
Oak Ridge National Laboratory	10 mg U	89.34% U 234
Carnegie Institution of Washington	0.7 mg U	99.94% U 235
Oak Ridge National Laboratory	10 mg U	99.94% U 235
Oak Ridge National Laboratory	10 mg U	99.9995% U 238
Argonne National Laboratory	1 gm U	99.9938% U 238
Knolls Atomic Power Laboratory	2.5 gm U	99.945 % U 238

ELECTROMAGNETIC FUNDAMENTALS

DC INJECTION OF IONS IN CYCLOTRONS

Preliminary tests of dc injection of protons in a cyclotron have indicated that the proton current increases as the dc potential applied to the accelerating electrode is increased. Since the dee voltage in the experimental unit was limited to 10 kv, application of injection potentials of over 10 kv resulted in deceleration of ions between the accelerating electrode and the dee. The equipment is now being redesigned to provide at least 20 kv dee-to-ground. Information obtained with the redesigned unit should then be more readily applicable to full-scale cyclotron operation.

ION SOURCE FOR MULTIPLY-CHARGED IONS

The hot-cathode, calutron-type ion source does not operate successfully at the high arc potentials required to obtain high outputs of multiply-charged ions. In an attempt to satisfy the conditions necessary for obtaining a copious emission of electrons, a grounded, gas-fed defining electrode has been inserted between the filament and the high voltage ionization chamber, Figure 13. The filament is maintained at $\sim$ 100 volts negative with respect to this defining electrode while a 1 to 20 kv positive potential is applied to the ionization chamber. A small quantity of gas supplied through the defining electrode tends to reduce space charge in the region of the filament and, by supplying positive ions to the filament, tends to increase emission. The deep defining slots at the ends of the ionization chamber are designed to confine the high-potential plasma; positive ions cannot readily flow from the chamber

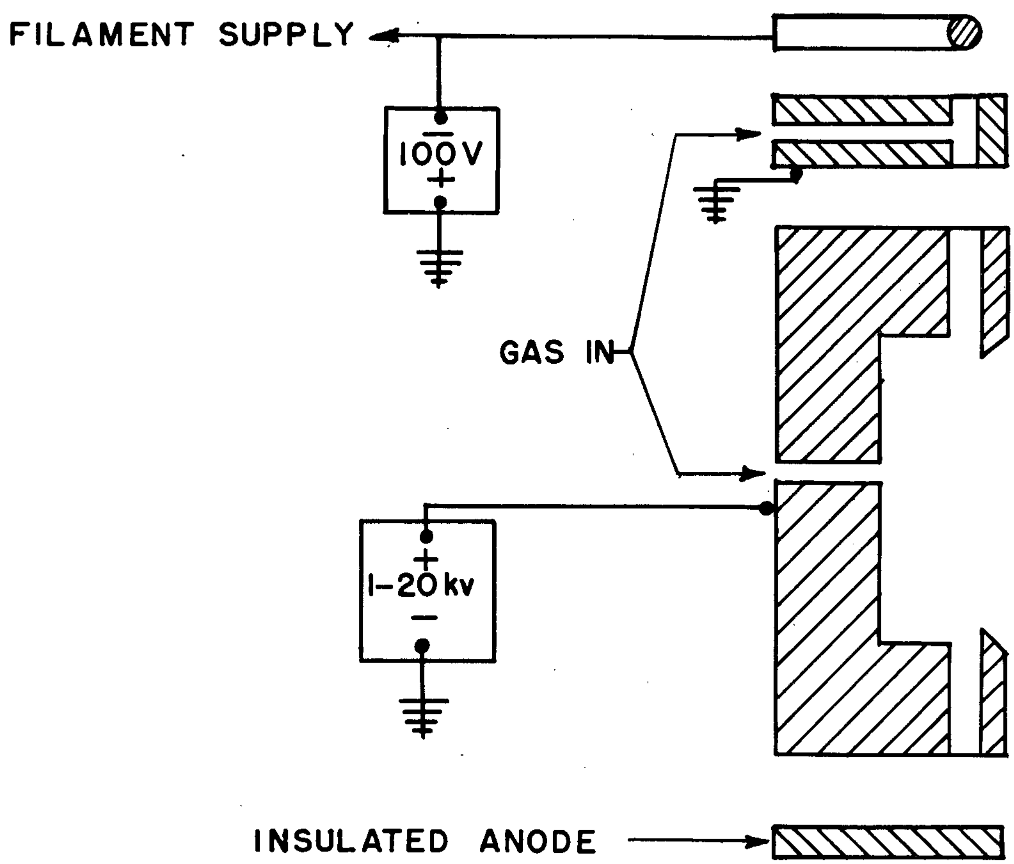

FIGURE 13 HIGH POTENTIAL ION SOURCE

[redacted]

to the cathode or anode. As a negative potential is built up on the insulated anode, electrons passing through the ionization chamber are reflected back into the chamber and continue to oscillate until potential gradients in the arc force them to the walls of the deep defining slots. At 10 kv it is estimated that primary electrons average 10 collisions before being lost to the wall of the chamber.

An experimental ion source of this type has been tested. The alignment of the electrodes with the magnetic field is critical. High energy electrons are obtained when no gas is fed to the chamber, but when gas is introduced the average electron energy is greatly reduced by the preponderance of low energy secondary electrons. To date it has not been possible to raise the energy of the arc sufficiently to produce a copious supply of multiply-charged ions.

[redacted]