AEC RESEARCH AND DEVELOPMENT REPORT

ORNL-1670
Progress

ELECTRONUCLEAR RESEARCH DIVISION
SEMIANNUAL PROGRESS REPORT
FOR PERIOD ENDING MARCH 20, 1954

OAK RIDGE NATIONAL LABORATORY
OPERATED BY
CARBIDE AND CARBON CHEMICALS COMPANY
A DIVISION OF UNION CARBIDE AND CARBON CORPORATION

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SEMIANNUAL PROGRESS REPORT
for Period Ending March 20, 1954

Robert S. Livingston, Director
Edited by F. T. Howard

DATE ISSUED

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OAK RIDGE NATIONAL LABORATORY
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PREVIOUS PROGRESS REPORTS

The following progress reports have been issued by the Electronuclear* Research Division for the indicated period-ending dates:

Y-495	September 30, 1949
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PAPERS PRESENTED DURING THE PERIOD

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B. L. Cohen, "Diffraction Scattering of Protons," Invited paper, American Physical Society meeting, New York, Jan. 28-30, 1954.

B. L. Cohen, W. H. Jones, G. H. McCormick, and B. L. Ferrell, "Angular Distribution of Fission Fragments from 22-Mev Proton-Induced Thorium Fission," American Physical Society meeting, New York, Jan. 28-30, 1954. Abstract: *Bull. Am. Phys. Soc.* **29**, 1 (1954).

E. Newman and W. L. Alford, "Angular Distribution of Alpha Particles from (p,α) Reactions," American Physical Society meeting, New York, Jan. 28-30, 1954. Abstract: *Bull. Am. Phys. Soc.* **29**, 1 (1954).

A. Zucker and H. L. Reynolds, "Excitation Functions for Nitrogen-Induced Nuclear Reactions in Carbon," American Physical Society meeting, New York, Jan. 28-30, 1954. Abstract: *Bull. Am. Phys. Soc.* **29**, 1 (1954).

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B. L. Cohen, "The Statistical Theory of Nuclear Reactions," *Phys. Rev.* **92**, 1245 (1953).

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W. R. Smith, "A Quick-Change Seal Opener," *Rev. Sci. Instr.* **24**, 886 (1953).

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B. L. Cohen, R. A. Charpie, T. H. Handley, and E. L. Olson, "Manganese-57," submitted to *The Physical Review*.

B. L. Cohen, W. H. Jones, G. H. McCormick, and B. L. Ferrell, "Angular Distributions of Fission Fragments from 22-Mev Proton-Induced Thorium Fission," submitted to *The Physical Review*.

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N. F. Ziegler, *An RF System for a Variable-Energy Cyclotron*, ORNL CF-54-2-92 (March 1, 1954).

A. L. Boch *et al.*, *A Ten-Megawatt Pressurized-Water Package Reactor and Associated Power Plant (Preliminary Draft)*, ORNL CF-53-10-106 (Oct. 1953).

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Part I

UNCLASSIFIED WORK

ELECTRONUCLEAR RESEARCH DIVISION SEMIANNUAL PROGRESS REPORT

ABSTRACT

Nuclear physics research with the 86-in. cyclotron included measurement of excitation functions and angular distributions of fission fragments for proton-induced fission in Th^{232}, U^{235}, and U^{238}; for several elements, the measurement of the angular distributions of alpha particles from (p,α) reactions, the excitation functions for $(p,2p)$ reactions, and the cross sections and excitation functions for $(p,2n)$ and (p,pn) reactions; and the identificotion of eight new isotopes. Thick-target yields for the production of eight isotopes of interest for medical and industrial application were determined in proton bombordments, usually exceeding 1 ma; in several cases the high yield and low bombardment costs suggest interesting production possibilities.

The 63-in. cyclotron now produces a deflected N^{+++} beam of 2 μa at approximately 29 Mev. The range-energy and the average charge-velocity relations of nitrogen ions in nickel were determined. The excitation functions for three nitrogen-induced reactions in carbon were then measured.

The design work for the revised 44-in. cyclotron is essentially completed and fabrication of new components is well under way.

An exploratory study has been completed and a proposal submitted for the construction of a 114-in. cyclotron for the acceleration of five nuclei, carbon through neon, to energies in excess of 100 Mev.

THE ORNL 86-INCH CYCLOTRON

NUCLEAR PHYSICS RESEARCH

Measurements were made of the angular distributions of fission fragments from 22-Mev proton-induced fission in Th^{232}, in U^{238}, and in U^{235}. Within the accuracy of the measurements, all angular distributions are symmetrical about the 90-deg angle and can be well fitted to an expression of the form:

$$I(\theta) = a + b \cos^2 \theta \ .$$

The values obtained for b/a are shown in Table 1.

Preliminary measurements for 15-Mev proton-induced fission indicate that angular distributions are not a sensitive function of incident-proton energy.

Measurements were made of the angular distribution of alpha particles from (p,α) reactions on nickel, niobium, and tantalum. Preliminary results indicate that the angular distributions have strong maxima in the forward direction, which suggests that (p,α) reactions proceed by direct as well as compound-nucleus interactions.

Excitation functions were measured for the $(p,2p)$ reactions in magnesium, calcium, iron, and zinc. They were found to be in reasonable agreement with the theoretical predictions.

Excitation functions were measured for proton-induced fission in Th^{232}, in U^{238}, and in U^{235}. In the uranium isotopes the cross sections are about equal to the total reaction cross sections at all energies. The cross section is only about half as large in Th^{232}. The U^{238} fission cross section is fitted by a nuclear radius of about $1.5\ A^{1/3} \times 10^{-13}$ cm.

Cross sections at 22 Mev were measured for several $(p,2n)$ and (p,pn) reactions in medium-weight elements; excitation functions were also measured in some cases. In all cases the (p,pn) cross section is considerably larger than the $(p,2n)$ cross section; this cannot be explained by any existing theory. The anomaly includes cases where each reaction is favored energetically and where intermediate and final nuclei of both reactions are even-even, odd-odd, even-odd, and odd-even. The sticking probability is considerably less than unity for elements as heavy as potassium ($Z = 19$) and probably for elements as heavy as iron ($Z = 26$).

TABLE 1. ANGULAR DISTRIBUTIONS OF FISSION FRAGMENTS

ISOTOPE	FISSION PRODUCT	MASS RATIO	b/a
Th^{232}	$Ag^{112,113}$	1.04	0.105 ± 0.025
	Ru^{105}	1.19	0.19 ± 0.08
	Zr^{97}	1.37	0.24 ± 0.025
	$Sr^{91,92}$	1.52	0.250 ± 0.020
	Ba^{139}	1.53	0.264 ± 0.015
U^{238}	$Ag^{112,113}$	1.10	0.09 ± 0.02
	Ba^{139}	1.43	0.15 ± 0.02
	Zr^{97}	1.43	0.18 ± 0.02
	$Sr^{91,92}$	1.58	0.17 ± 0.04
	Br^{83}	1.94	0.16 ± 0.04
U^{235}	$Ag^{112,113}$	1.07	0.105 ± 0.02
	Zr^{97}	1.40	0.16 ± 0.02
	$Sr^{91,92}$	1.55	0.19 ± 0.02
	Br^{83}	1.81	0.12 ± 0.04

Attempts to measure the (p,He^3) cross section for gallium indicated that it is less than the theoretical predictions by at least a factor of 10. This is the first case where charged-particle emission was found to be less probable than was predicted from theory.

Eight previously unknown isotopes were identified and are listed in Table 2.

APPLIED PHYSICS

Members of the Applied Physics Group determined the thick-target yield for several isotopes of interest to industry and the medical profession. Other work included a study of the ion-transmission characteristics of the cyclotron at very low energy, and a preliminary test of the feasibility of accelerating multiply charged heavy ions.

Radioisotope Production

Several long runs were made during this period for the quantity production of radioisotopes. Since no facilities for radiochemical work are available, the chemical phase of the program is performed by the Radioisotope Development Department of the Operations Division. Distribution of isotopes produced in the 86-in. cyclotron is handled by the Radioisotopes Sales Department.

Cobalt-56

In a 38-hr run for the production of 80-d Co^{56} by the $Fe^{56}(p,n)Co^{56}$ reaction, the estimated yield was 100 mc/mahr. The activity produced during this bombardment is being used for gamma-ray scattering studies at the Radiation Laboratory of the University of California, Berkeley. Cobalt-56 is of interest in this work because it emits a large percentage of gamma rays with unusually high energy. Its gamma-ray energies are $\gamma 1 = 0.845$, $\gamma 2 = 1.26$, $\gamma 3 = 1.74$, $\gamma 4 = 2.01$, $\gamma 5 = 2.55$, and $\gamma 6 = 3.25$; they are emitted in the ratio

$$\gamma 1/\gamma 2/\gamma 3/\gamma 4/\gamma 5/\gamma 6 = 1.0/0.5/0.2/0.1/0.2/0.2.$$[1]

The Co^{56} yield, after chemical processing, was 2.2 curies or 75% of the expected yield. The low yield was apparently caused by poor bonding of the iron coating to the silver-plated aluminum target base; the iron had been applied with a metallizing gun. In spite of sparking difficulties, an average current of 800 μa was obtained for

[1] L. G. Elliot and M. Deutsch, *Phys. Rev.* **64**, 321 (1943).

TABLE 2. NEW RADIONUCLIDES IDENTIFIED

RADIONUCLIDE	HALF LIFE	RADIATION	METHOD OF PRODUCTION	METHOD OF IDENTIFICATION
Er^{161}	3.6 h	X ray and 0.184-, 0.823-, and 1.10-Mev gammas	$Er^{162}(p,pn)Er^{161}$, $Er^{162}(p,2n)Tm^{161}$ with decay to Er^{161}	Not produced by p on Ho or (n,p) on Er
Ho^{161}	2.5 h	X ray and 0.90-Mev gamma	Proton bombardment with Er with decay of Er^{161} to Ho^{161} or $Dy^{161}(p,n)Ho^{161}$	Daughter of Er^{161}, milking of Er^{161} to obtain Ho^{161}
Ho^{167}	3.0 h	1.0 ± 0.1 Mev β^- and 0.35-Mev gamma	$Er^{170}(p,\alpha)Ho^{167}$, $Er^{167}(n,p)Ho^{167}$	Excitation
Pr^{136}	70 m	X ray, β^+, and 0.17-Mev gamma	Enriched $Ce^{136}(p,n)Pr^{136}$	Excitation
Pr^{135}	22 m	X ray, β^+, and 0.080-, 0.22-, and 0.30-Mev gammas	Enriched $Ce^{136}(p,2n)Pr^{135}$	Parent of Ce^{135}
Pr^{137}	1.3 h	β^+	Enriched $Ce^{138}(p,2n)Pr^{137}$	
$Pr^?$	13 m	β^+	Proton bombardment of Ce isotopes	
Mn^{57}	1.7 m	2.6-Mev β^- and 0.117-, 0.131-, 0.69-, 0.35(?)-, and 0.22(?)-Mev gammas	Enriched $Fe^{57}(n,p)$	Cross section, comparison with other isotopes, comparison of gamma energies with those of Co^{57}

the run. Inspection of the target after removal from the cyclotron indicated that the iron had been sputtered from about 2 sq in. of target surface. During a subsequent shutdown period the interior of the cyclotron, especially the dee tips, was found to be quite radioactive (~25 r/hr on contact). The radioactive material consisted of loosely adherent particles of iron, most of which was removed by vacuum cleaning. Rough estimates indicate that as much as 100 mc was removed. In the future, iron coatings will be applied to Co^{56} production targets by electrodeposition to assure better bonding.

Cobalt-57

Cobalt-57 is produced by the (p,pn) and $(p,2n)$ reactions on Ni^{58}. Because of its relatively long half life (270 d) and soft gamma rays (principally 120 kv), Co^{57} should be suitable for industrial radiography of low-density metals and alloys or thin sections of the heavier metals. A survey of the literature indicates that no entirely suitable reactor-produced isotope is available for the range between Ir^{192} and beta rays. A recent bombardment of 68%-abundant Ni^{58} has shown that Co^{57} is produced at the rate of 50 mc/hr at a proton-beam current of 1 ma. The cost of bombardment is then about $1/mc, which seems low enough to make applications requiring curie quantities of Co^{57} entirely feasible.

Sodium-22

Sodium-22 is usually produced in cyclotrons by the (d,α) reaction on Mg^{24} (79% natural abundance). With high-energy protons, Na^{22} is produced by the (p,α) reaction on 10.1% Mg^{25} and the $(p,\alpha n)$ reaction on 11.1% Mg^{26}; at energies below 22 Mev most of the yield is due to the former reaction. Yield calculations based on published cross sections[2] give a thick-target yield of 0.4 mc/mahr. Although the yield from 20-Mev deuterons is higher, deuteron machines usually produce a somewhat smaller current than does the 86-in. cyclotron; consequently, the production rates are nearly equal. The economics are such that an error of 10 to 20% in the published cross sections could easily make production of carrier-free Na^{22} at ORNL attractive.

During a 12.9-hr test run to determine the actual yield of Na^{22}, an average current of 1150 μa was maintained on a target fabricated entirely of 98% Mg–2% Al alloy. The target was of the grazing-incidence type, 6 × 10 in., with water passages formed by rifle drilling $^3/_{16}$-in. holes its full length. Cooling water was supplied at an inlet pressure of 235 psig. A coolant flow of 100 gpm was obtained, producing a mean water velocity in the cooling passages of 50 fps. The yield from this run has not yet been determined.

Sodium-22 can also be produced by the (p,pn) reaction on sodium metal. The exploitation of this reaction for the large-scale production of Na^{22} is complicated by the low melting point of the sodium and the undesirability of using sodium compounds because of their low thermal conductivity. Calculations show that, with high-velocity water cooling and with attention to minimizing the thickness of metal through which heat must be transferred, large beam currents can be used without exceeding the melting point of the surface of the sodium target.

In the first attempt to bombard a metallic sodium target, an average proton current of 200 μa was maintained for 5 hr; a Na^{22} yield of 4.46 mc/mahr was obtained. In a more recent test a sodium-coated rifle-drilled copper-base target was bombarded continuously for 48.5 hr at an average current of 410 μa. The estimated yield for this test is 90 mc, and the specific activity should approach 10 mc per gram of sodium.

Manganese-54 (Carrier-Free)

Carrier-free Mn^{54} is produced in the 86-in. cyclotron by the (p,n) reaction on 2.4% Cr^{54}. The yield is 0.5 mc/mahr as determined by a run of 9.4-hr duration. In a subsequent run an average current of 1020 μa was maintained on a conventional 6 × 10 in. grazing-incident target with slotted water passages.

In bombarding normal chromium, the presence of Cr^{52} (83.7%) leads to the production of very large quantities of undesirable activity. Manganese-52 is produced by the (p,n) reaction at the rate of about 1.6 curies/mahr, and Cr^{51} by the (p,pn) reaction at about 600 mc/mahr. Large quantities of Mn^{52} and Cr^{51} lead to considerable difficulty in removing the target from the cyclotron and complicate chemical processing. Three days after completion of a 24-hr bombardment, the radiation level was still about 25 r/hr at a distance of 5 ft from the target.

The use of separated isotopes is being seriously considered in an attempt to reduce the production of the undesirable activities and to increase the rate of Mn^{54} production.

Iron-55 and Manganese-54 (Non-Carrier-Free)

Proton bombardment of manganese produces both Mn^{54}, by the (p,pn) reaction, and Fe^{55}, by the (p,n) reaction. The estimated yields for Mn^{54} and Fe^{55} are 33.4 and 9.7 mc/mahr, respectively.

Since the Fe^{55} is produced carrier-free at a relatively low cost (~$5/mc), it may replace reactor-produced Fe^{55}. Iron-55 with high specific activity is presently produced in the reactor by the (n,γ) reaction on calutron-separated Fe^{54}. The price of such material is $50/mc. Another advantage of the cyclotron product is that it is completely free of Fe^{59}.

The Mn^{54} produced by the $Mn(p,pn)$ reaction is not carrier-free; consequently it has more limited application. Specific activities in the 50- to 100-mc/g range seem entirely feasible, and because the cost is so much lower than that of carrier-free Mn^{54} ($1.50/mc vs $100/mc), it is anticipated that considerable demand may develop.

The manganese target was prepared by electroplating manganese directly onto a 6 × 10 in. curved aluminum base by using a process suggested by representatives of the Electro-Manganese Corporation. In a 7-hr test, an average current of 1170 μa was maintained with no evidence of target deterioration.

Arsenic-73,74

Arsenic isotopes ($As^{73,74}$) are produced by the (p,n) and $(p,2n)$ reactions on germanium. Germanium is a difficult metal to fabricate because

[2]R. E. Batzel and G. H. Coleman, *Cross Sections for Formation of Na*^{22} *from Aluminum and Magnesium Bombarded with Protons*, MTA-27 (Jan. 15, 1953).

of its high melting point, hardness, and brittleness; therefore the capsule irradiation target was used. Two grams of powdered germanium metal was sealed in a thin-walled Inconel capsule and irradiated at a current of 200 μa for 5 hr. The 17.5-d As^{74} was produced at a rate of 10 mc/hr, and the 76-d As^{73} at 1.6 mc/hr.

Yttrium-88

Proton bombardment of Sr^{88} (natural abundance 83%) produces 105-d Y^{88} by the (p,n) reaction. Because of its high melting point and reactivity with oxygen, strontium metal targets are difficult to fabricate. Consequently, strontium oxide was irradiated in the thin-walled capsule target. Approximately 21 mc of Y^{88} was produced in a 5-hr test run at a current of 200 μa. The estimated yield is 52 mc/mahr at 100% target efficiency, indicating that in this case the target efficiency was 50% after correction was made for the use of the oxide rather than the metal.

Gallium-67

The production of Ga^{67} for the ORINS Medical Division was continued at the rate of 240 mc on alternate weeks. This 68-h isotope is used mostly in tracer experiments.

Other Isotopes

Numerous short bombardments were made for the production of small quantities of various activities for other ORNL divisions and for off-site institutions. These included bombardment of chromium for Mn^{52}; cesium for Ba^{133}; thallium for the several lead isotopes; bismuth for Po^{207}; lead for the various bismuth isotopes; gold for Hg^{195} and Hg^{197}; tantalum for W^{181}; ruthenium for Rh^{102}; and calutron-separated Cd^{111} for In^{111}.

Ion-Transmission Tests

Several tests were made with the 86-in. cyclotron adjusted for the transmission of ions, heavy ions as well as protons, at resonance modes other than the first. Normally the machine is tuned for the first mode; that is, during each half-cycle of applied radio frequency the protons transit 180 deg of their orbit. Ions requiring 3, 5, 7, etc. half-cycles to transit 180 deg can also be accelerated in a two-dee cyclotron. The cyclotron is tuned for these modes by adjusting the magnetic field.

If the radio frequency applied to the cyclotron dees is held constant, the ratio of resonance magnetic field for a given ion to the field normally used for a proton is

$$\frac{H}{H_p} = \frac{M}{nk} ,$$

where M is the mass of the ion relative to the mass of a proton; k is the charge number of the ion; and n is the mode number, with the restriction that n must be an odd integer. The ratio of the energy of the ion to the energy of a proton for the same radius is, then,

$$\frac{E}{E_p} = \frac{M}{n^2} .$$

With the magnetic field of the 86-in. cyclotron reduced to one-third, protons were accelerated on the third mode. Currents up to 30 ma were measured calorimetrically on the target at 25 in. from the center of the magnetic field. The energy of the protons is approximately 1 Mev under these conditions. At this output the ion-transport efficiency (target current divided by the sum of target current and ion loss to the dees) was approximately 70%. The insulation of the dee system from ground makes it possible to measure the dee drain, or loss of ions to the dees.

Triply charged nitrogen ions were also accelerated by operating the cyclotron for resonance at the fifth mode for N^{14}. In the first preliminary experiment, 30 μa of 12-Mev N^{+++} ions was obtained on a target 30.5 in. from the center of the magnet. It should be pointed out that at this same magnetic field N^{5+} could be accelerated at the third mode. Acceleration of triply charged N^{14} at the third mode would require a higher magnet current than the magnet cooling system was designed to accommodate.

Operation as a Neutron Source

The ORNL Biology Division is currently using the cyclotron about 4 hr per week in studies of the effect of fast neutrons on biological materials. In these experiments the neutrons are produced by bombarding a thick beryllium target with 22-Mev protons. Test organisms are irradiated in a lead box with 1-in. walls which is used to reduce the gamma-ray background. The center of the box is 52 in. from the target, in the forward direction of the beam. Neutrons entering the lead chamber pass through 2 in. of stainless steel in addition to the 1-in. lead wall. The neutron flux and dose rate at the center of the chamber were recently measured by the Health Physics Division. The preliminary data gave a flux density of 1.6×10^9 neutrons/sq cm·sec and a dose rate of 3.7 rep/sec at a target current of 1 ma.

BEAM DEFLECTOR DEVELOPMENT

A beam-deflection system is being developed for the 86-in. cyclotron to make it possible to deflect protons from the cyclotron dees to external targets placed in a separate reaction room. Both an electrostatic deflector and a magnetic channel (weakened region in the field) are required.

Iron "bars" used to weaken the magnetic field in a certain region for the deflection of the beam also weaken and disturb the magnetic field over a large portion of the circulating beam. A coil wound around the iron and energized by a current flowing in such a direction as to further weaken the magnetic field in the beam channel produces, however, an external field which strengthens the magnetic field in the circulating-beam region. Much of the disturbance of the magnetic field in the circulating-beam region can be corrected with such a coil. An edge-wound 32-turn coil of 1-in. copper ribbon, 0.25 in. thick, will be energized at currents up to 5000 amp. The coil will be cooled by water flowing through a $\frac{3}{16}$-in.-OD tube which is silver-soldered to the edge of the copper conductor. A test section of conductor, cooled in this manner, was operated at 40,000 amp/sq in. without an excessive temperature rise.

The water-corrosion test of aluminum tubing (0.050-in. OD and 0.003-in. wall) for use in the deflector septum was terminated at 2000 hr. The tube then weighed 5% more than its original weight. The tubing was also tested in cyclotron beams up to 1500 μa. The longest test before failure was 30 hr at 540-μa average beam current. The principal cause of failures has been leaks at the joints where the tubes bond to the header. This is apparently due to corrosion caused by flux remaining in the solder joint. A further difficulty has been that a large portion of the tubing, as received from the manufacturer, contained holes through the wall. Thicker-walled tubing was ordered from a different manufacturer.

Engineering drawings have been completed for the dees, trimmer capacitors, liner, deflector and adjustment mechanism, and the reaction room. The dees are being fabricated by an outside contractor. Many associated components, being fabricated locally, are nearly completed.

CYCLOTRON OPERATION

The 86-in. cyclotron has been operated routinely for the past several months. The dees were removed for servicing once because of a sparking condition. It was found that a number of screws had been loosened as much as $\frac{1}{4}$ in.; a few had fallen out. Apparently, mechanical vibration is a problem to be considered in the design of cyclotron dees. Cyclotron operation has been uneventful since the dees were serviced.

The work schedule has continued at two shifts a day, five days a week, with an occasional third shift being required for a long continuous run. During the past six months the following bombardments have been made:

Nuclear physics research	391
Applied physics (isotope production)	65
Cyclotron development	29
Neutron production for Biology Division (~375 exposures)	27
Operational tests	16
Total	528

The average innage, exclusive of the one-week shutdown for dee servicing, was 43.6% or 34.9 hr of actual operation per 80-hr week. The large number of short runs, many of only 1-min duration, resulted in low innage. The time summary for a typical week is as follows:

	Hours	Per Cent
Innage (beam on target)	32.2	40.2
Attempted innage and bakeout time	8.5	10.6
Source – routine change of filament and adjustments	2.0	2.5
Targets – routine change and inspection	21.6	27.0
Equipment failure and troubles	2.7	3.4
Standby (no target available)	9.3	11.6
Radiation hazard	1.5	2.8
Weekly startup and shutdown	2.2	1.9

The use of Octoil-S oil in the diffusion pumps has proved to be unsatisfactory. A gummy mass accumulates on the pump surfaces and interferes with the pumping action. It has been necessary to dismantle the 8-in. diffusion pumps for cleaning about every six months. So far, the 20-in. pumps have not required cleaning. When the cyclotron is shut down for the installation of the beam deflector, the large pumps will be dismantled, cleaned, and refilled with DP-20 diffusion-pump oil.

THE 63-INCH CYCLOTRON

The 63-in. cyclotron now produces a deflected beam of about 2 μa, available for experimental purposes at the exit port. The energy of these deflected N^{+++} ions was measured to be 29.1 ± 0.3 Mev.

Many nuclear-reaction experiments involving nitrogen ions require the use of thin foils either as energy attenuators or as windows in gas chambers. Also, knowledge of the range-energy relations for nitrogen ions is essential if thick-target yields are to be interpreted as cross sections. For these two reasons it was necessary to determine the range-energy curves of nitrogen ions. Previous experimental work in the field was inconclusive because no source of energetic heavy ions was available.

The salient feature of the penetration of matter by heavy particles is the process of capture and loss of orbital electrons by the ions. Since this process is not well understood, measurements were made of the average charge of nitrogen ions as a function of their velocity in nickel, along with the range of nitrogen ions in nickel. Once the range-energy relations were established, excitation functions were measured for three reactions in carbon and one in beryllium.

The range-energy relations of nitrogen ions in nickel were measured by bombarding hydrogen gas with the cyclotron beam and measuring the range of proton recoils at 0 deg in nuclear emulsions. The energy of the nitrogen ions was varied by interposing suitable nickel absorbers in the beam. The range-energy curve obtained for nitrogen ions in nickel is shown in Fig. 1. It should be noted that the energy loss is approximately constant over the range measured.

The N^{+++} beam was directed through a collimator into a Faraday cup used to measure the beam current. Nickel foils of various thickness were placed in the beam path, and changes in the charge of the nitrogen ions were then determined by changes in the current collected by the Faraday cup. From these data the curve for average ion charge vs absorber thickness was obtained (Fig. 2), and with this curve and the range-energy relation it was then possible to determine the average charge of the nitrogen ions as a function of velocity (energy). The results are given in Fig. 3.

Excitation functions for the following reactions were measured:

$$C^{12}(N^{14})F^{18}Be^{8}$$
$$C^{12}(N^{14})Na^{24}2H^{1}$$
$$C^{12}(N^{14})Na^{22}He^{4}$$
$$Be^{9}(N^{14})F^{18}He^{4}n$$

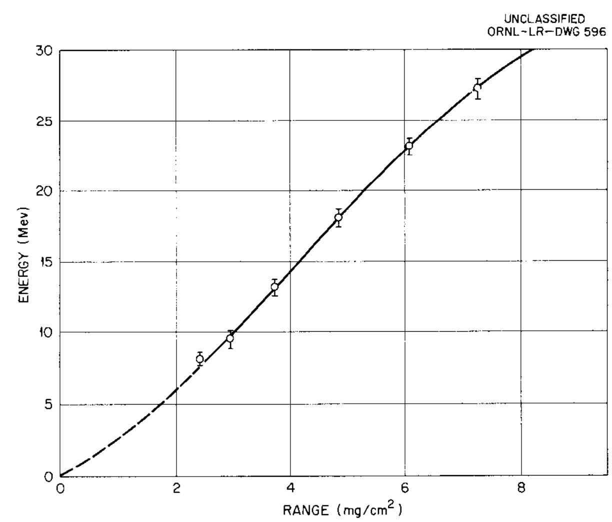

Fig. 1. Range-Energy Relation of Nitrogen Ions in Nickel.

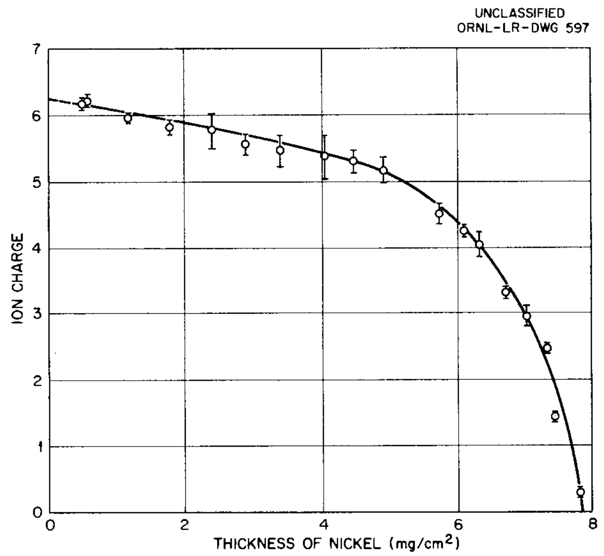

Fig. 2. Charge of Nitrogen Ions vs Thickness of Nickel Absorbers.

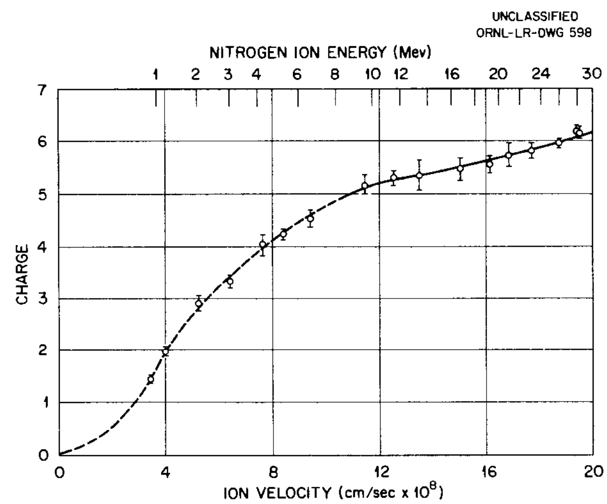

Fig. 3. Charge of Nitrogen Ions vs their Velocity in Nickel.

The cross sections of the reactions with carbon as a function of energy are given in Fig. 4. These reactions were observed from the radioactive residual nuclei, and the cross sections were measured with absolute beta counting.

An article interpreting the physical significance of these results is being submitted for publication in *The Physical Review.*

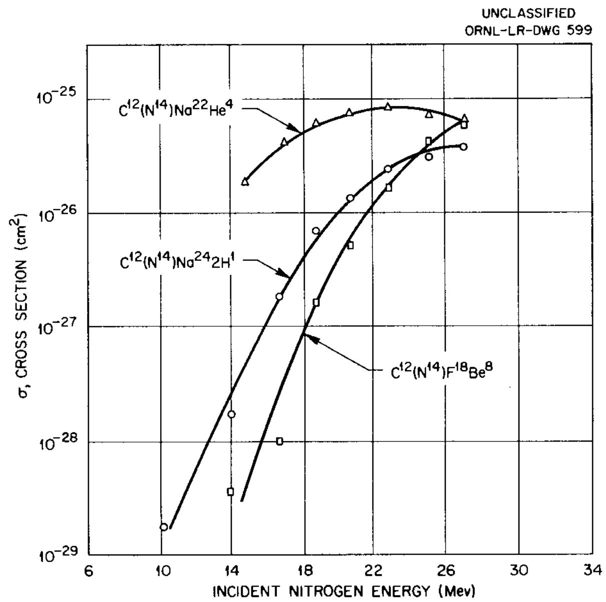

Fig. 4. Excitation Functions for Nitrogen-Induced Reactions with Carbon.

THE 44-INCH CYCLOTRON

Design work for the revision of the 44-in. cyclotron is essentially completed; the new components are being fabricated and assembled. The liner, dees, and dee-stem housing are being fabricated by an outside contractor. The faceplates, dee stems, ion source, target probe, and vacuum system were fabricated locally and are being assembled.

To provide for greater flexibility in the development program planned for this cyclotron, the magnet pole faces (tank walls) are being ground with a portable grinder to provide a very uniform magnetic field, as near ± 0.01% as possible. This will then provide a standard base for the various magnetic shim designs that may be tested.

A detailed drawing has been prepared showing the 44-in. cyclotron completely assembled (Fig. 5). In this figure the dees are shown in the position to be used in accelerating protons to approximately 5 Mev. In the cross-section drawing, Fig. 6, the 14.5-in. spacer is shown in place between the main faceplate and the vacuum tank. This spacer moves the dees back from the center of the magnetic field so that the machine can be operated at approximately 1.5 Mev for test operation at very high proton currents.

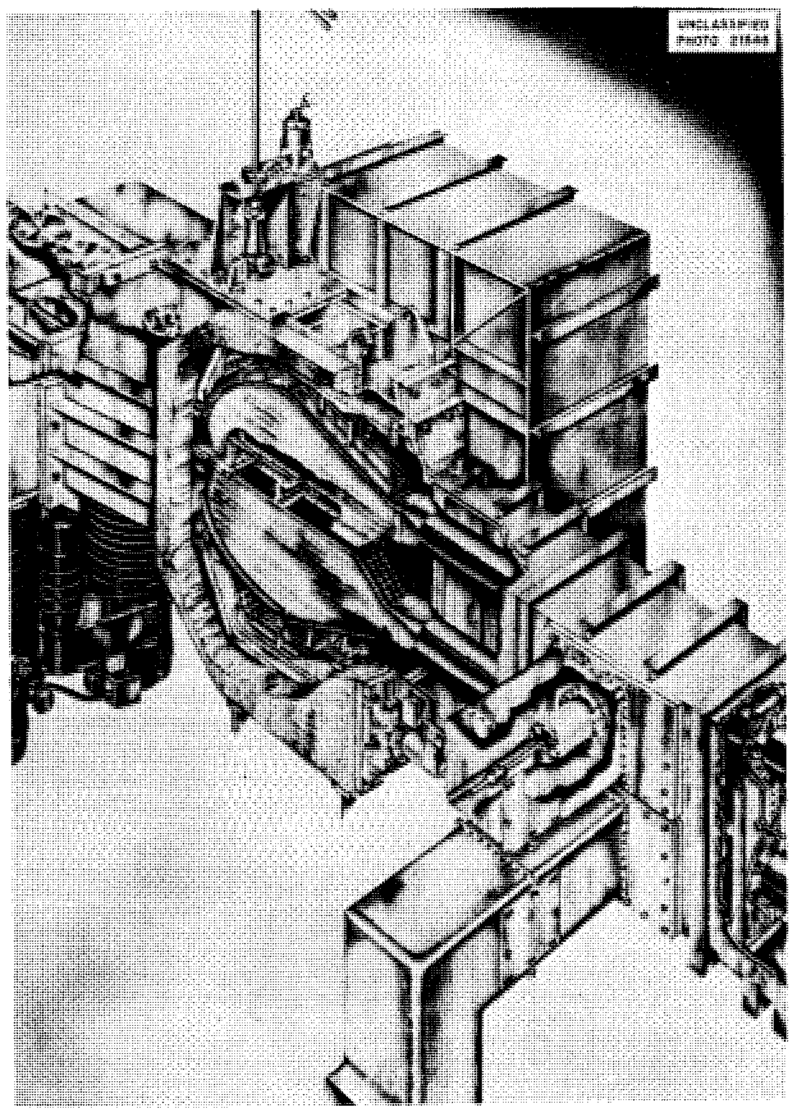

Fig. 5. ORNL 44-Inch Cyclotron.

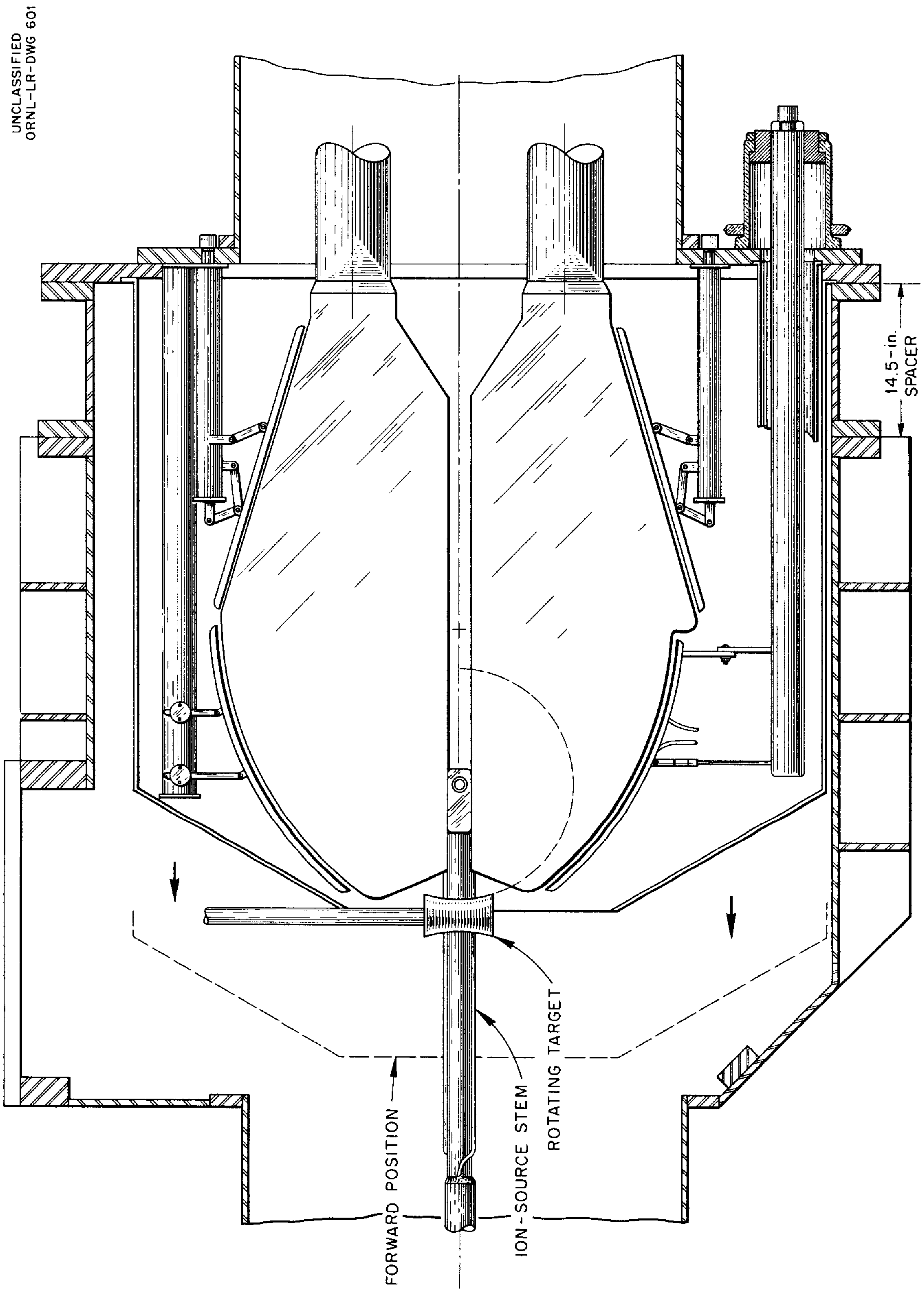

Fig. 6. Cross Section of 44-Inch Cyclotron With Spacer in Place for Operating at Reduced Energy.

MULTIPLE-PARTICLE HEAVY-ION CYCLOTRON

The successful operation of the 63-in. cyclotron for the acceleration of N^{+++} to 25 Mev and the production of many new types of reactions that are obtained with heavy particles have stimulated wide interest in the construction of a larger heavy-particle accelerator which would be capable of producing nuclear reactions in even the heaviest elements. An exploratory study was made at this Laboratory for the construction of a fixed-frequency, high-current cyclotron designed to accelerate multiply charged carbon, nitrogen, oxygen, fluorine, and neon ions to energies in excess of 100 Mev.

The proposed machine combines some of the design features of both the 86- and 63-in. cyclotrons. It resembles the former in the high voltages needed for acceleration and the general size of the machinery. It resembles the 63-in. cyclotron insofar as the low frequency, the magnetic field, and the source of multiply charged ions are concerned. By holding the magnetic field constant and varying the radio frequency, it would be possible to accelerate ions of five elements. The particles and their characteristics are listed in Table 3.

The proposed machine requires a magnet with a pole-base diameter of 114 in. and a field of 20,000 oersteds in a clear gap of 11.5 in. The total weight of the magnet is approximately 980 tons.

The radio-frequency resonant system, Fig. 7, consists of conventional dees which are excited from and suspended by two vertical dee stems, a shorting plate, a variable condenser, and a liner. A grounded-grid type of oscillator circuit furnishes radio-frequency power to the dee system.

In connection with the study of a multiple-particle heavy-ion cyclotron, an investigation of a low-pass filter coupling to cyclotron dee stems was completed. In this experiment a low-pass prototype "T" having a characteristic impedance of 500 ohms was connected to the lower dee stem of the 63-in. cyclotron model. The resonant frequency of the dee stems was varied with vacuum variable capacitors connected from the end of each dee stem to ground. Data concerning the operation of the filter as a function of frequency indicate that a low-pass filter could be used to couple the oscillator to the dee stems of a variable-frequency system, provided a variable phase-shift circuit were included in the grid circuit of the oscillator.

Also, in connection with the study of a multiple-particle cyclotron, an automatic tuning system was developed which should prove applicable to such a machine. The tuning system operates very reliably in tuning a small oscillator to the resonant frequency of a loosely coupled secondary circuit. In operation the automatic control sweeps the

TABLE 3. ACCELERATED PARTICLES AND THEIR CHARACTERISTICS

ION	ATOMIC NUMBER	MASS	ENERGY (Mev)		ENERGY OF BARRIER** (Mev)
			Kinetic	COM System*	
C^{3+}	6	12	133	127	78
N^{3+}	7	14	114	108	90
N^{4+}	7	14	203	192	90
O^{3+}	8	16	100	94	101
O^{4+}	8	16	178	167	101
F^{4+}	9	19	150	139	112
Ne^{4+}	10	20	142	131	124
Ne^{5+}	10	20	222	205	124

*Center-of-mass system, on U^{238}.

**Coulomb barrier for penetration of U^{238}.

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Photo 21830

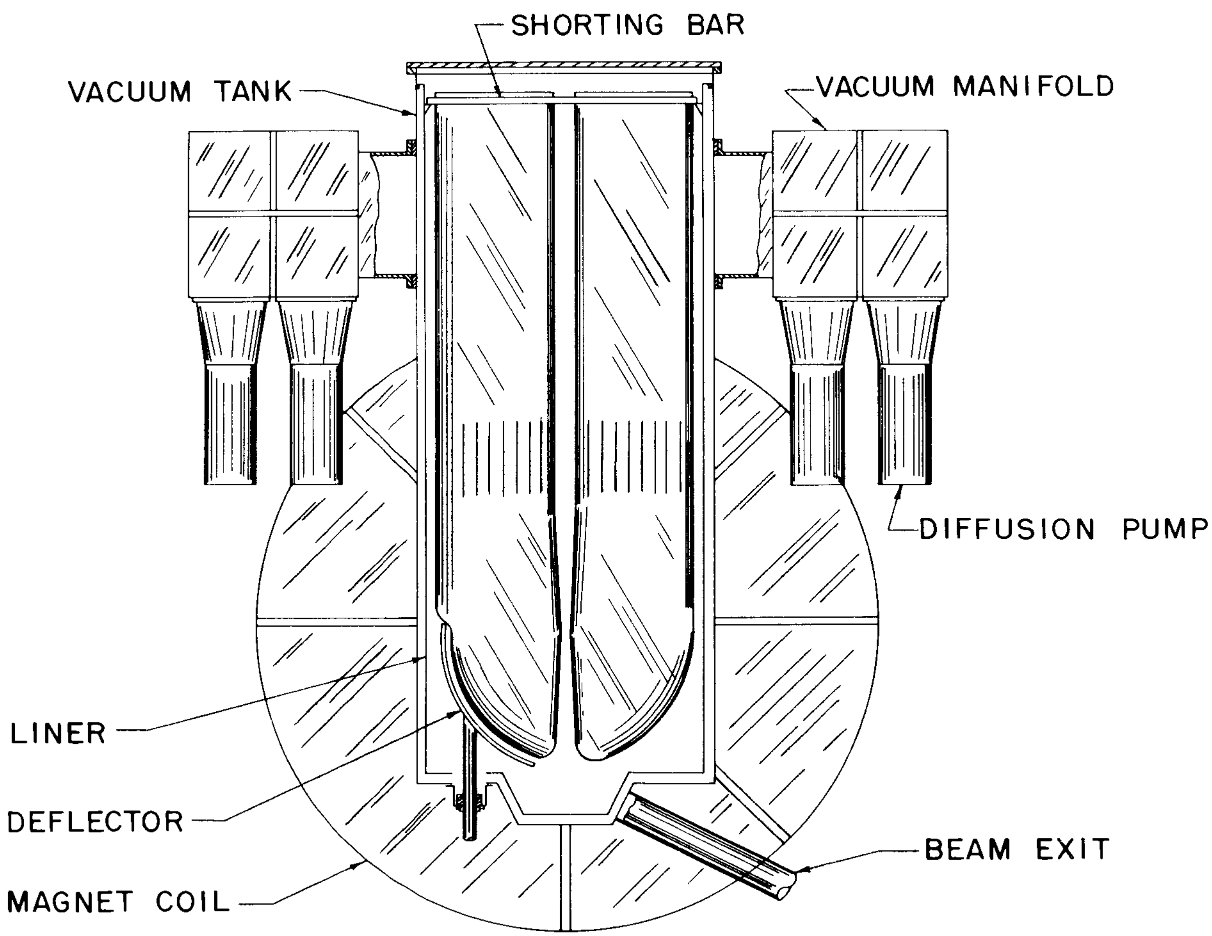


Fig. 7. Section through Proposed 114-Inch Multiple-Particle Heavy-Ion Cyclotron.

oscillator frequency through its entire range and "stores" the maximum voltage developed across the tuned secondary on a capacitor. The control again causes the oscillator frequency to sweep through its range and during this second sweep compares the voltage developed across the secondary with the "stored" voltage on the capacitor. When the two voltages are equal the sweep is stopped, leaving the oscillator tuned to the same frequency as that of the secondary circuit. The error of the control in tuning the oscillator to the frequency of the secondary is of the order of 0.1%. This is thought to be within the accuracy required of the multiple-particle cyclotron.

Part II

[redacted] WORK

ABSTRACT

The first two runs for the enrichment of gram quantities of plutonium isotopes were successfully completed; the first run yielded 0.5 g of 36% Pu^{240}. In thorium separation, 3.3 g of Th^{230} (ionium) was obtained. Techniques for confining the alpha-active materials being processed were proved to be satisfactory.

The preliminary feasibility and design report for a 10-Mw pressurized package reactor was completed and issued. Further design details were worked out for the final report being prepared for submission, by the AEC, with invitations for lump-sum contracts for construction of the prototype of this reactor.

SPECIAL SEPARATIONS

PLUTONIUM PROGRAM

Plutonium isotope separation was undertaken for the production of small quantities of enriched isotopes for use in measurements of nuclear properties and for the evaluation of the electromagnetic process for large-scale separations for special weapon application. The feed bank for the project is of two levels of irradiation: 477 g assaying 5.6% Pu^{240} and 140 g assaying 12.8% Pu^{240}.

Calutron performance was determined for a run charged with 140 g of plutonium assaying 5.6% Pu^{240} and 0.36% Pu^{241}. The calutron equipment included a two-arc ion-source unit and three-pocket collectors for masses 239, 240, and 241. The radius of the ion orbits in this 180-deg mass spectrograph is approximately 24 in., and the one-mass-difference separation of the peaks at the collector is only 0.10 in. The plutonium weight and isotopic analysis of the three products obtained from this first run are given in Table 4, and the isotopic enrichment factors (enhancement) and process efficiencies are shown in Table 5.

A second run for the separation of isotopes from the 5.6% Pu^{240} feed has been completed, but the products have not yet been assayed. After one more run at this feed level, some 12.8% Pu^{240} will be processed for the high enrichment of the heavier isotopes.

The distribution of electromagnetically enriched isotopes of heavy elements to various laboratories since September 1953 is given in Table 6.

THORIUM-230 PROGRAM

Plutonium isotope separation facilities and methods were pretested with thorium isotope

TABLE 5. PLUTONIUM CALUTRON PERFORMANCE

PRODUCT	ENRICHMENT FACTOR	PROCESS EFFICIENCY (%)
Pu^{239}	239:240 = 9	5.7
Pu^{240}	240:239 = 32	5.2
Pu^{241}	241:239 = 57	5.2

TABLE 4. PLUTONIUM ISOTOPES COLLECTED

COLLECTOR POCKET	ISOTOPIC COMPOSITION (at. %)			WEIGHT OF PLUTONIUM (g)
	Pu^{239}	Pu^{240}	Pu^{241}	
Pu^{239}	99.39	0.60		7.11
Pu^{240}	36.5	62.8	0.6	0.55
Pu^{241}	65.9	19.7	14.3	0.17

TABLE 6. DISTRIBUTION OF ISOTOPES

ISOTOPE	TO	AMOUNT (mg)	PURITY (%)
U^{234}	Brookhaven National Laboratory	1,689	13.43
U^{235}	Brookhaven National Laboratory	1,693	99.86
	Metals and Controls Corporation	2,032	99.86
	National Bureau of Standards	843	99.86
	Oak Ridge Notional Laboratory, Y-12	8,470	99.50
	Oak Ridge National Laboratory, X-10	5,870	99.50
	Naval Research Laboratory	9,400	97.34
U^{236}	Brookhaven National Laboratory	4	94.90
	Oak Ridge National Laboratory, X-10	15	94.90
U^{238}	Phillips Petroleum Company	17	99.99992
	Metals and Controls Corporation	25,400	99.9988
	Oak Ridge National Laboratory, X-10	850	99.9956
	Knolls Atomic Power Laboratory	1,016	99.9956
	General Electric Research Laboratory	18,270	99.992
	Brookhaven National Laboratory	1,023	99.992
	Argonne National Laboratory	187,370	99.968
	Brookhaven National Laboratory	549,800	99.947
	Oak Ridge National Laboratory, X-10	4,234	99.946
Th^{230}	Carnegie Institution of Washington	1	90.61
	California Institute of Technology	18	90.02
	Los Alamos Scientific Laboratory	211	90.02
	Oak Ridge National Laboratory, X-10	27	90.02
Pu^{240}	Argonne National Laboratory	9 5	12.88

separation runs in which the feed was a batch of thorium containing 0.6 curie of Th^{230} (ionium). Thorium-230 is an alpha emitter of approximately one-third the specific activity of Pu^{239}. Argonne National Laboratory supplied 294 g of thorium as an oxide of isotopic ratio of Th^{232}:Th^{230} = 9:1. This material was produced by extraction from Mallinckrodt raffinate.

The electromagnetic separation of the thorium isotopes was both for the preparation of Th^{230} of maximum isotopic purity and for testing plutonium equipment and unit-transfer techniques. A total of 3.3 g of 90% Th^{230} was obtained. Gram quantities of purified Th^{230} are to be used for the production of high-purity Pa^{231} and U^{232} by neutron irradiation in the MTR. Thorium feed was run through the complete cycle twice so that the plutonium wash area and laboratory equipment could be thoroughly tested. At the same time, transfer of units and disassembly of the calutron could be checked for dispersal of alpha activity. It was found that the use of Vinylite enclosures when transferring units of calutron equipment successfully confined the alpha-active thorium and minimized the health hazard.

The chemical cycle was patterned after the analagous process for plutonium and included the following principal phases: (1) vacuum scraping of metal deposits from graphite collector pockets and subsequent ignition of product scrapings to the oxide form and (2) the recovery of unresolved residues from calutron surfaces and the purification and conversion of recovered residues to the chloride. The last of the material is now being processed.

PACKAGE REACTOR PROJECT

The preliminary feasibility and design report of a 10-Mw pressurized package reactor and associated power plant was completed and issued.[1] This plant, rated at 1000 kw of electricity and 3535 kw of steam for space heating, is proposed for military use at a remote arctic installation. The objectives of the study program were

1. to make a feasibility study, including sufficient design data to arrive at a fairly accurate cost estimate for the plant,
2. to make use of proved techniques and methods in the design so that the development work required could be held to a minimum,
3. to serve as a basis for further economic studies,
4. to provide a relatively firm proposal from which private contractors could be expected to submit construction bids.

Preliminary letters have been sent (by the AEC) to a number of private companies to solicit information on their interest in undertaking the design and construction of a prototype plant based on the ORNL feasibility report. It is anticipated that a lump-sum contract for construction of the plant will be requested. Several phases of the reactor design are currently being investigated.

FUEL-PLATE FABRICATION

For the past several months members of the Metallurgy Division have been engaged in fuel-plate fabrication development work on the flat stainless-steel-clad fuel elements for the reactor. To date, several complete subassemblies have been successfully clad and brazed. No serious difficulties are expected in the fabrication of a complete core for the reactor. A summary of this work will be found in the progress report of the Metallurgy Division.

CONTROL-ROD DRIVE MECHANISM

A contract has been granted to the American Machine and Foundry Company of New York City for the construction and testing of a control-rod drive mechanism, as proposed for the package reactor. The tests will be rather detailed and extensive in that all actual conditions, with the exception of radiation, will be simulated. The complete control rod, including dummy fuel plates, and drive mechanism will be constructed to scale. The system will be operated under actual conditions of temperature, pressure, and water circulation; it will be cycled for a sufficiently long period of time to establish reliability, wear effects, corrosion effects, and other pertinent characteristics. Initial testing is expected to commence about April 1, 1954.

REACTOR SIMULATOR TESTS

The package reactor has been examined by means of the ORNL reactor power plant simulator for stability resulting from changes in load and reactivity. The results of the program[2] indicate good stability when temperature coefficients of greater than 2×10^{-5} are used. Reactivity steps, up to 0.5%, were introduced without the temperature of the fuel or the coolant exceeding established maximum values. Drastic changes in load conditions did not result in untenable effects.

PLANT-TESTING PROCEDURES

A complete set of plant-testing procedures is being written. These procedures will be used as part of the plant specifications when prospective contractors are invited to submit bids.

PRIMARY-LOOP HYDROGENATION

The need for primary-loop degassification is being reinvestigated. This is particularly pertinent to the problem of primary-loop hydrogenation. Evidence obtained from the STR Project at Arco indicates that degassification is neither necessary nor desirable when the reactor is operating with hydrogenation. It is used only when the system is depressurized during startups or shutdowns.

CORROSION STUDIES

The information accumulated since 1948 on the corrosion of potential reactor materials is being analyzed as a preliminary to the preparation of specifications for the reactor. The power reactor is composed of two basic components: the reactor

[1] A. L. Boch, W. R. Gall, and W. H. Jordan – F. T. Howard (ed.), *A Ten-Megawatt Pressurized-Water Package Reactor and Associated Power Plant. (Preliminary Draft)*, ORNL CF-53-10-106 (Oct. 1953).

[2] E. R. Mann and F. P. Green, *Analog Simulation in the Package Reactor Study*, ORNL CF-54-1-104 (Mar. 2, 1954).

itself with the primary loop; and the power plant with its associated equipment, which might be considered the secondary loop. Present concern is with the primary loop since this loop contains the potentially radioactive liquids and is consequently the most probable source of trouble.

Although the solubility of any material which is to be used in contact with water is a function of its mechanical properties and its wear resistance, its corrosion resistance is by far the dominating factor in the present application. The mechanism of corrosion is generally accepted as an electrochemical process in a continually changing equilibrium. As a matter of convenience, factors which may affect corrosion have been listed as follows:

1. Dissolved gases (O_2, H_2, CO_2, N_2, degassed)
2. Welding, heat treatment, sensitizing
3. Fluid flow
4. Irradiation of materials
5. Irradiation of water – its dissociation and recombination
6. Transport-corrosion materials – their formation and deposition
7. Stress
8. Water purity
9. pH
10. Inhibitors and additives
11. Galvanic couples and crevices
12. Surface finish
13. Acid cleaning and passivation

The results of the many experiments run at Hanford, Argonne, Battelle, ORNL, Babcock and Wilcox, Westinghouse, General Electric, and others are being tabulated according to the above factors. This tabulation has progressed far enough so that it can be stated that specification of materials and water conditions can be based upon experience with these reactors.

NUCLEAR CALCULATIONS

The nuclear calculations which were made for the preliminary engineering design of the package reactor have been critically reviewed. These calculations were based on two-group diffusion theory and are considered to yield insufficiently accurate results for final design purposes. A method has been devised for multigroup solution of the Boltzmann equation in *P*-3 approximation, for spherical hydrogenous reactors. The method takes into account the correlation between neutron energy loss and scattering angle in collisions with hydrogen. It provides also for a thin absorbing shell between core and reflector. Coding of this method for solution in the ORACLE will be undertaken shortly. Interim calculations on the UNIVAC indicate that nearly half the fissions in the fully loaded reactor are caused by neutrons in the intermediate-energy range.

A detailed analysis of fission-product poisons has been made by taking into account all known fission products and the best cross sections available for them. For the flux levels and time scale encountered in the package reactor, the macroscopic cross sections of all the important poisons except xenon and samarium depend on the time-integrated flux.

Progress is currently being made toward reliable estimates of critical mass, control-rod effectiveness, and flux distributions. It is expected that these results will be available for the revised report.

The preliminary feasibility and design report[1] is being revised and amplified to include the available results from the above investigations. It is expected that the revised report (ORNL-1613) will be ready for issuing by the time the invitations to bid on the plant are sent to the prospective contractors.

THE APOTRON

Because of increased emphasis on other projects, the investigation of the apotron for the separation of light isotopes has been discontinued. During this program, knowledge of the control of intense gaseous discharges was increased, new high levels of ion output were achieved, improved methods of dissipating ion-beam power were tested, and other engineering features pertinent to the operation of large-scale ion sources were explored.

In experimental operation of the apotron with lithium chloride as feed, ion currents as high as 3.5 amp were obtained from a single arc. During runs for production tests, stable operation was maintained with ion currents up to 2.5 amp for a single-arc unit and with total ion currents up to 6.2 amp for a three-arc unit. The enrichment factor for Li^6 varied from an average of 18.5 to a maximum of 28; for Li^7 it varied from 24 to 27. The process efficiency, varying somewhat from run to run according to the equipment used, averaged 35% with a maximum of 55%. In this work, primary emphasis was on increasing the output of the ion source. Both the enrichment factor and the process efficiency could be improved considerably by giving more attention to the design of the ion collector.

OPERATIONS SUMMARY

A summary of operations in Buildings 9204-3 and 9201-2 from September 20, 1953 to March 20, 1954 includes the following experimental runs:

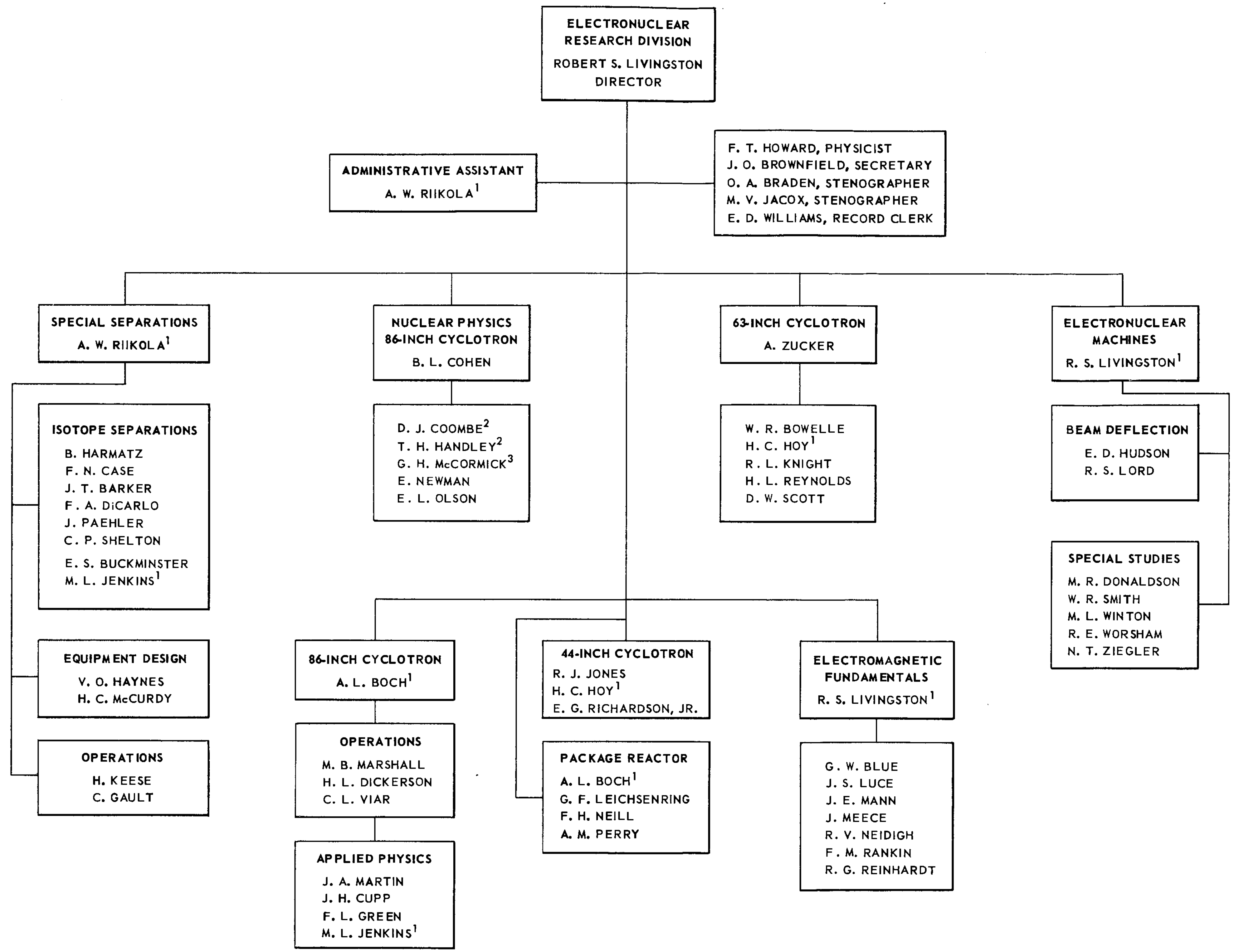
Special separations	26
Electronuclear fundamentals	64
Electronuclear machines	10
The 86-in. cyclotron	528
The 63-in. cyclotron	68
The apotron	57

The distribution of shop time charged to each of the various projects is given in Table 7.

Assignment of personnel to the various projects is indicated in the organization chart.

TABLE 7. TIME DISTRIBUTION – MECHANICAL SERVICE AND MACHINE SHOPS, SEPTEMBER 20, 1953 TO MARCH 20, 1954

ACCOUNT NUMBER AND PROJECT	MECHANICAL SERVICE		MACHINE SHOP			
			Carbon		Metal	
	Hours	Per Cent of Total Time	Hours	Per Cent of Total Time	Hours	Per Cent of Total Time
33 Special separations	3,150.6	16.7	1158.0	60.2	429.0	13.1
34 86-in. cyclotron, applied physics	1,111.0	5.9	30.0	1.6	98.0	3.0
35 86-in. cyclotron, operations	4,348.2	23.1	7.0	0.4	581.6	17.7
36 Fundamental electronuclear research	2,918.0	15.5	321.0	16.7	87.0	2.6
37 Electronuclear machines	119.0	0.6			127.0	3.9
39 86-in. cyclotron, nuclear physics	75.0	0.4			24.0	0.7
40 63-in. cyclotron, physics	189.0	1.0	22.0	1.1	156.0	4.7
41 63-in. cyclotron, operations	1,751.5	9.3	91.0	4.7	651.0	19.8
42 Apotron development	2,798.5	14.9	292.0	15.2	36.0	1.1
43 Package reactor						
44 44-in. cyclotron	2,342.8	12.4	2.0	0.1	1097.4	33.4
1 Building maintenance	32.0	0.2				
Total	18,835.6	100	1923.0	100	3287.0	100



[1]Dual Capacity
[2]On Laon from Analytical Chemistry Division
[3]Research Participant

Personnel: Monthly 41
Weekly 12
53