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SEMIANNUAL PROGRESS REPORT

FOR PERIOD ENDING NOVEMBER 20, 1955

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SEMIANNUAL PROGRESS REPORT

For Period Ending November 20, 1955

C. P. Keim, Director

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G. E. Boyd, J. R. Sites, Q. V. Larson, and C. R. Baldock, "Production and Identification of Long-Lived Technetium Isotopes at Masses 97, 98, and 99," *Phys. Rev.* **99**, 1030 (1955).

H. W. Dunn, *A Study of X-Ray Fluorescence for the Analysis of Rare Earths and Other Complex Groups*, ORNL-1917 (July 13, 1955).

E. L. Grove and J. A. Norris, "A Study of Matrix Effects on a Graphite Base Semi-Quantitative Procedure," *Appl. Spectroscopy* **9**, 63 (1955).

C. E. Melton, L. O. Gilpatrick, Russell Baldock, and R. M. Healy, *Improved Techniques for the Isotopic Analysis of Boron on the Mass Spectrometer*, ORNL-2006 (November 1955).

G. L. Rogosa and W. F. Peed, *U L_I Transition from Separated Isotopes*, ORNL-2009 (November 1955).

Boyd Weaver, *Chemical Refinement Procedures in the Electromagnetic Separation of Isotopes*, ORNL-1952 (September 1955).

O. B. Rudolph, *Relay Insulation Studies*, ORNL-1916 (July 12, 1955).

H. M. Rosenstock, A. L. Wahrhaftig, and H. Eyring, "Metastable Ions in Mass Spectra," *J. Chem. Phys.* **23**, 2220 (1955).

H. M. Rosenstock, J. R. Sites, J. R. Walton, and Russell Baldock, "The Mass Spectra of CuCl, CuBr, CuI," *J. Chem. Phys.* **23**, 2442 (1955).

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ASTM Committee E-14 on Mass Spectrometry, San Francisco, California, May 22–27

Russell Baldock, J. K. Gladden, and L. O. Gilpatrick, "Considerations of Isotope Dilution Analysis."

C. P. Keim and Russell Baldock, "The Large Electromagnetic Isotope Separators at Oak Ridge."

Ohio State Symposium on Molecular Structure and Spectroscopy, Ohio State University, Columbus, June 13–17

H. W. Morgan and J. H. Goldstein, "The Microwave Spectrum and Molecular Structure of Vinyl Fluoride."

Gordon Research Conference on the Chemistry and Physics of Isotopes, Meriden, New Hampshire, July 10–16

P. S. Baker, "Utilization of Electromagnetically Enriched Isotopes."

Russell Baldock, "Current Status of Isotope Abundance Measurements."

L. O. Love, "Stable Isotope Separating Facilities at the Oak Ridge National Laboratory."

J. R. Sites, "Techniques for Isotopic Abundance Measurements of the Platinum Metals."

Boyd Weaver, "Chemistry in the Electromagnetic Separation of Isotopes."

Gordon Research Conference on Inorganic Chemistry, New Hampton, New Hampshire, July 11–15

Boyd Weaver, "Rare-Earth Separations at the Oak Ridge National Laboratory."

Conference on the Production and Utilization of Electromagnetically Enriched Isotopes, Harwell, England, September 13–16

C. P. Keim and C. R. Baldock, "Mass Spectrometry at the Oak Ridge National Laboratory."

C. P. Keim and P. S. Baker, "Distribution and Utilization of Electromagnetically Enriched Isotopes from 1946 to 1955 (U.S.A.)."

C. E. Normand, L. O. Love, W. A. Bell, Jr., and W. K. Prater, "High-Temperature Ion Sources."

C. E. Normand, L. O. Love, and W. A. Bell, Jr., "Isotope Collector Developments."

C. E. Normand, "The Oak Ridge National Laboratory Isotope Separators."

Boyd Weaver, "Preparation of Charge Materials for Electromagnetic Separation of Isotopes."

Boyd Weaver, "Chemical Refinement of Electromagnetically Separated Isotopes."

Meeting of the Optical Society of America, Pittsburgh, Pennsylvania, October 6–8

P. M. Griffin and K. L. Vander Sluis, "The Application of Electrodeless Discharge Sources and the Echelle Spectrograph to Zeeman Spectroscopy."

J. R. McNally, Jr., "High Resolution Spectroscopy at the Oak Ridge National Laboratory."

K. L. Vander Sluis and J. R. McNally, Jr., "The Fabry-Perot Interferometer with Finite Apertures."

Meetings of the Tulsa, Kansas City, and St. Louis Sections of the Instrument Society of America, October 3, 4, and 5, respectively

J. R. McNally, Jr., "Principles and Applications of Instrumentation in Spectroscopy, Isotope Research."

Conference on Rare Earths in Biochemical and Medical Research, Oak Ridge Institute of Nuclear Studies, October 27–29

Boyd Weaver, "Separation of Rare Earths by Liquid-Liquid Extraction."

OTHER MEETINGS ATTENDED BY MEMBERS OF THE DIVISION

C. E. Normand and C. P. Keim, Gordon Research Conference on the Chemistry and Physics of Isotopes, July 11–16, 1955.

O. C. Yonts, Eighth Annual Gaseous Electronics Conference, Schenectady, New York, October 20–22, 1955.

W. F. Peed, Fourth Annual Symposium of Industrial Application of X-Ray Analyses, University of Denver, Denver, Colorado, August 11–12, 1955.

PAPERS CONCERNING USAGE OF ENRICHED ISOTOPES

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L. Koerts, P. Macklin, B. Farrelly, R. Van Lieshout, and C. S. Wu (Columbia University), "Radiations of I-126," *Phys. Rev.* **98**, 1230–1237 (1955).	Te-126
H. Mark, C. McClelland, and C. Goodman (MIT), "Electric Excitation of Certain Medium-Weight Nuclei by Protons," *Phys. Rev.* **98**, 1245–1251 (1955).	Cr-53; Pd-105; Cd-111, -113
G. M. Temmer and N. P. Heydenburg (Carnegie Institution of Washington), "Properties of Excited States of Pd and Cd Nuclei by Coulomb Excitation," *Phys. Rev.* **98**, 1308–1309 (1955).	Pd-108, -110; Cd-116
A. O. Burford, J. F. Perkins, and S. K. Haynes (Vanderbilt University), "Relative Intensities of the Radiations from Hf-175," *Phys. Rev.* **99**, 3–7 (1955).	Hf-174
J. A. Harvey, D. J. Hughes, R. S. Carter, and V. E. Pilcher (BNL), "Spacings and Neutron Widths of Nuclear Energy Levels," *Phys. Rev.* **99**, 10–33 (1955).	Sn(8 isotopes); Mo-95, -96, -97; In-113, -115
P. H. Stelson and F. K. McGowan (ORNL), "Gamma-Ray Yields from Coulomb Excitation," *Phys. Rev.* **99**, 112–127 (1955).	Pb-206
R. K. Sheline and J. R. Wilkinson (Florida State University), "Isobaric Triplet Cr-48–V-48–Ti-48," *Phys. Rev.* **99**, 165–169 (1955).	Ti-46
P. B. Sogo and C. D. Jefferies (University of California, Berkeley), "Spin, Magnetic Moment, and Electric Quadrupole Moment of La-138, and the Magnetic Moment of Cl-36," *Phys. Rev.* **99**, 613 (1955).	La-138
P. H. Stelson and F. K. McGowan (ORNL), "Coulomb Excitation of States in Thallium and Lead," *Phys. Rev.* **99**, 616 (1955).	Pb-206
G. Paulissen and H. Mark (MIT), "Electric Excitation of Palladium, Cadmium, and Molybdenum by Protons," *Phys. Rev.* **99**, 617 (1955).	Cd-113, -114; Pd-105, -106, -108, -110
G. M. Temmer and N. P. Heydenburg (Carnegie Institution of Washington), "Energy Levels in Ru, Pd, and Cd by Coulomb Excitation," *Phys. Rev.* **99**, 617 (1955).	Pd-104, -105, -106, -108, -110; Cd-110, -111, -112, -113, -114, -116
N. P. Heydenburg and G. M. Temmer (Carnegie Institution of Washington), "Energy Levels in Ti, Fe, Ge, and Mo by Coulomb Excitation," *Phys. Rev.* **99**, 617 (1955).	Mo-98, -100
L. A. Toller, J. R. Patterson, and H. W. Newson (Duke University), "Neutron Resonances in the Kilovolt Region: Cl-35, Cl-37, K-39, and K-41," *Phys. Rev.* **99**, 620 (1955).	Cl-35, -37; K-39, -41
P. F. Nichols, R. G. Smith, and H. W. Newson (Duke University), "Total Neutron Cross Sections in the Kilovolt Region: Zr-90, Zr-92, Zr-94," *Phys. Rev.* **99**, 621 (1955).	Zr-90, -92, -94
F. Ajzenberg, A. Rubin (Boston University), and J. G. Likely (Princeton), "Neutrons from the Proton Bombardment of S-32 and S-34," *Phys. Rev.* **99**, 654 (1955).	S-32, -34
C. K. Bockelman, C. M. Braams, W. W. Buechner, and D. B. Guthe (MIT), "Angular Distribution of Proton Groups from Ca-42(d,p)Ca-43 Reaction," *Phys. Rev.* **99**, 655 (1955).	Ca-42
G. C. Phillips, C. R. Gossett, J. P. Schiffer, and P. M. Windham (Rice Institute), "Inelastic Scattering of Protons from Fe-54 and Fe-56," *Phys. Rev.* **99**, 655 (1955).	Fe-54, -56

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J. P. Schiffer, P. M. Windham, C. R. Gossett, and G. C. Phillips (Rice Institute), "Inelastic Scattering of Protons from Isotopes of Mn, Ni, and Cu," *Phys. Rev.* **99**, 655 (1955).	Mn-55; Ni-58, -60; Cu-63, -65
C. E. Weller and J. C. Grosskreutz (University of Texas), "Gamma Rays from the Proton Bombardment of Cu-63 and Cu-65," *Phys. Rev.* **99**, 655 (1955).	Cu-63, -65
D. W. Martin, J. M. Cork, and S. B. Burson (ANL), "Radiations from Ce-143(33 hr)," *Phys. Rev.* **99**, 670 (1955).	Ce-142
B. L. Cohen, E. Newman, and T. H. Handley (ORNL), "(p,pn) + $(p,2n)$ and $(p,2p)$ Cross Sections in Medium Weight Elements," *Phys. Rev.* **99**, 723–728 (1955).	Zn-68; Si-29
J. R. Wilkinson and R. K. Sheline (Florida State University), "New Isotope Manganese-53," *Phys. Rev.* **99**, 752 (1955).	Cr-53
G. E. Boyd, J. R. Sites, Q. V. Larson, and C. R. Baldock (ORNL), "Production and Identification of Long-Lived Technetium Isotopes at Masses 97, 98, and 99," *Phys. Rev.* **99**, 1030–1031 (1955).	Mo-98
R. M. Sinclair (Westinghouse Research Laboratories), "Gamma Radiation from the Interaction of 4.4-Mev Neutrons with Fe-54 and Fe-56," *Phys. Rev.* **99**, 1351–1353 (1955).	Fe-54
L. L. Lee, Jr. and W. Rall (Yale University), "Angular Distributions for Ti-46, 48(d,p)Ti-47, 49 Reactions with 4.16-Mev Deuterons," *Phys. Rev.* **99**, 1384–1387 (1955).	Ti-46, -48
J. D. Kington, J. K. Bair, H. O. Cohn, and H. B. Willard (ORNL), "Proton-Neutron Threshold Measurements," *Phys. Rev.* **99**, 1393–1397 (1955).	Mg-25; Cu-63
T. H. Handley and W. S. Lyon (ORNL), "Neutron-Deficient Activities of Terbium," *Phys. Rev.* **99**, 1415–1418 (1955).	Gd-152, -154, -155, -156, -157, -158
B. Farrelly, L. Koerts, N. Benczer, R. Van Lieshout, and C. S. Wu (Columbia University), "Radiations from Sb-122," *Phys. Rev.* **99**, 1440–1446 (1955).	Sb-121
G. M. Temmer and N. P. Heydenburg (Carnegie Institution of Washington), "Fast E2 Transition Probabilities from Coulomb Excitation," *Phys. Rev.* **99**, 1609–1612 (1955).	25 different samples
J. B. Cumming (BNL), "A New Zinc Isotope, Zn-61," *Phys. Rev.* **99**, 1645 (1955).	Ni-58
R. H. Hughes (University of Wisconsin), "Isotope Shift in the First Spectrum of Atomic Lithium," *Phys. Rev.* **99**, 1837–1839 (1955).	Li-6, -7
E. W. Burke, Jr. (University of Wisconsin), "Isotope Shift in the First Three Spectra of Boron," *Phys. Rev.* **99**, 1839–1842 (1955).	B-10, -11
J. K. Bair, J. D. Kington, and H. B. Willard (ORNL), "Gamma-Ray and Neutron Yields from the Proton Bombardment of Boron," *Phys. Rev.* **100**, 21 (1955).	B-10
R. M. Brugger, T. W. Bonner, and J. B. Marion (Rice Institute), "Study of the Nuclear Reactions Sc-45(p,n)Ti-45, Cu-63(p,n)Zn-63, Cu-65(p,n)Zn-65, and Zn(p,n)Ga," *Phys. Rev.* **100**, 84–91 (1955).	Cu-63, -65
N. P. Heydenburg and G. M. Temmer (Carnegie Institution of Washington), "Coulomb Excitation of Rare-Earth Nuclei with Alpha Particles," *Phys. Rev.* **100**, 150–167 (1955).	Nd-143, -145; Sm-147, -148, -149, -150; Hf-176, -177, -179; Gd-155

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A. R. Brosi and B. H. Ketelle (ORNL), "Radioactivity of the Cerium-137 Isomers," *Phys. Rev.* **100**, 169–171 (1955).	Ce-136
J. M. Cork, M. K. Brice, L. C. Schmid, and R. G. Helmer (University of Michigan), "Radioactive Decay of Ruthenium-97," *Phys. Rev.* **100**, 188–191 (1955).	Ru-96
B. L. Cohen (ORNL), "(p,γ) Cross Sections," *Phys. Rev.* **100**, 206–209 (1955).	Fe-54; Ni-60
H. Mark and G. T. Paulissen (MIT), "Electric Excitation of Certain Rare-Earth Nuclei by Protons," *Phys. Rev.* **100**, 813–823 (1955).	Sm-148, -150, -152, -154; Gd-154, -156, -158, -160
G. R. Harrison, R. C. Crawford, and J. I. Hopkins (Vanderbilt University), "K-Series Fluorescence Yields of Argon, Copper-65, and Indium-113," *Phys. Rev.* **100**, 841–845 (1955).	Sn-112
J. B. Marion, T. W. Bonner, and C. F. Cook (Rice Institute), "(d,n) Reactions on Boron and Carbon," *Phys. Rev.* **100**, 847–851 (1955).	B-10
J. M. Cork, M. K. Brice, L. C. Schmid, and R. G. Helmer (University of Michigan), "Gamma Energies in Tc-97 Following K Capture in Ru-97," *Phys. Rev.* **100**, 955 (1955).	Ru-96
F. Ajzenberg, A. Rubin, and G. D. Johnson (Boston University), "Neutrons from the Proton Bombardment of B-11," *Phys. Rev.* **100**, 960 (1955).	B-11
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F. A. Jenkins and D. R. Speck (University of California), "Nuclear Moments of Gadolinium from Optical Hyperfine Structure," *Phys. Rev.* **100**, 973 (1955).	Ga-155, -157
K. R. Osborn and H. E. Gunning (Illinois Institute of Technology), "Determination of Hg-202 and Other Mercury Isotopes in Samples of Mercury Vapor by Mercury Resonance Radiation Absorbiometry," *J. Opt. Soc. Am.* **45**, 552 (1955).	Hg-202
I. C. Gardner (National Bureau of Standards), "Light Waves and Length Standards," *J. Opt. Soc. Am.* **45**, 685 (1955).	Cd-114
P. M. Griffin and K. L. Vander Sluis (ORNL), "Application of Electrodeless Discharge Sources and the Echelle Spectrograph to Zeeman Spectroscopy," *J. Opt. Soc. Am.* **45**, 901 (1955).	Te-130; Hg-202
J. R. McNally, Jr. (ORNL), "High Resolution Spectroscopy at Oak Ridge National Laboratory," *J. Opt. Soc. Am.* **45**, 901 (1955).	Li-6, -7
K. Burns (University of Pittsburgh, Allegheny Observatory), "Precise Wavelengths in the Spectra of Isotopes," *J. Opt. Soc. Am.* **45**, 903 (1955).	Cd-114

STABLE ISOTOPES RESEARCH AND PRODUCTION DIVISION SEMIANNUAL PROGRESS REPORT

SPECTROSCOPY RESEARCH LABORATORY

J. R. McNally, Jr., Department Head

INTRODUCTION

The Spectroscopy Research Laboratory is concerned with research and development in the fields of nuclear magnetic resonance, microwaves, infrared and optical spectroscopy, spectrochemistry, and x rays. Research and development are directed toward the determination of fundamental spectroscopic data and the investigation of methods of analysis for isotopes, elements, and compounds.

Outstanding accomplishments during this semiannual period are as follows:

1. The nuclear magnetic resonances of enriched Ba^{135} and Ba^{137} have been measured (Project NR-2).
2. Anharmonicity corrections to infrared data on T_2O, THO, and TDO result in verification of Libby's theoretical calculations (Project IR-2).
3. Zeeman studies of tellurium are being made to a precision of ± 0.0025 g units for individual Zeeman patterns (Project OS-1).
4. An expression for correcting Landé g factors for the anomalous magnetic moment of the electron is given (Project OS-3).
5. A spectrochemical method using yttrium as a carrier has been developed for below the part per million levels of rare earths in metals (Project SC-1).
6. Precision x-ray measurements of technetium, plutonium, americium, and neptunium have been obtained (Project X-2).

NUCLEAR RESONANCE

Project NR-1 – Nuclear Magnetic-Resonance Equipment

H. E. Walchli O. B. Rudolph

An a-c chopper amplifier was installed in the nuclear magnetic-resonance magnet regulator. Also, a new voltage-scanning source was constructed for use with the new regulator. Field regulation appears to be about 1 part in 100,000.

Experiments have been performed on an absorption-type spectrometer for the purpose of measuring the high strength field in the magnet used for Zeeman spectroscopic studies (see Project OS-1). The magnet, which has 1-in.-dia poles and a $\frac{1}{4}$-in. gap, does not seem to have a sufficiently homogeneous field for visual presentation of the deuterium resonance. Additional experiments are to be performed.

Project NR-2 – Nuclear Magnetic Resonances and Chemical Shifts

H. E. Walchli T. J. Rowland[1]

In collaboration with T. J. Rowland, the electron diamagnetic shielding effect (Knight shift) in finely ground, normal cadmium metal was measured (at the Metals Research Laboratory of Electro-Metallurgical Company) relative to the enriched stable isotope Cd^{113} in the form of Cd(I). The shift $\Delta H/H$ was found to be 0.0042.

Measurements of the nuclear magnetic resonances of Ba^{135} and Ba^{137} were performed. The results of these measurements, on enriched stable isotopes of barium in the form of saturated aqueous $BaCl_3$ in magnetic fields of approximately 9250 and 8300 gauss, gave

$$\nu_{Ba^{135}}/\nu_{Cl^{35}} = 1.01387 \pm 0.00002 ,$$

$$\nu_{Ba^{137}}/\nu_{Cl^{35}} = 1.13420 \pm 0.00005 ,$$

$$\nu_{Ba^{137}}/\nu_{Ba^{135}} = 1.11868 \pm 0.00006 .$$

A previous measurement based on a single observation at ORNL[2] gave $\nu_{Ba^{135}}/\nu_{Cl^{35}}$ as 1.014 ± 0.001. The increased sensitivity due to the higher resolution of Rowland's instrument permitted more adequate observations of the barium and cadmium

[1]Metals Research Laboratory of Electro-Metallurgical Company, Niagara Falls, New York.

[2]H. E. Walchli, *Stable Isotope Research and Production Semiann. Prog. Rep. Nov. 20, 1954*, ORNL-1829, p 1.

resonances than had been possible with the older ORNL spectrometer.

Measurements were also made in the diamagnetic shielding in five boron organic complexes and in boric acid, with B^{11} being used in both solutions. The interpretation of the results is not yet complete.

MICROWAVE SPECTROSCOPY

Project M-1 – Microwave Spectrum of Vinyl Fluoride

H. W. Morgan J. H. Goldstein[3]

The experimental work in the study of the microwave spectrum of vinyl fluoride has been completed. A summary of the project is now being written for publication and will be submitted to *The Journal of Chemical Physics*.

Measurements of the frequencies of several transitions in a Q-type series ($\Delta J = 0$) failed to yield a more precise value of the rotational constant a. Thus there still exists an uncertainty of ±0.011 A in the C=C bond length of 1.359 A determined from the data previously reported.

Project M-2 – Microwave Spectrum of Vinyl Acetylene

H. W. Morgan J. H. Goldstein

A least-squares procedure for nonlinear equations has been coded for the Oracle by the Mathematics Panel and will be employed to determine the best values of the various parameters from the microwave data previously reported.

Project M-3 – Microwave Spectra of Tritium Compounds

H. W. Morgan J. R. Lawson[4]
J. H. Goldstein

The microwave spectrometer and its associated vacuum system have been adapted for the handling of tritium compounds.

The first molecule chosen for study was TCN, since its microwave transition frequencies could be predicted with some accuracy. Two preparations of this compound were made and were studied in the spectrometer. A very rapid exchange rate made it impossible to verify the TCN transitions or to make precise measurements.

The source of the rapid tritium exchange has been studied, since the rate is appreciably faster than the rates that occur for samples of DCN under identical conditions. It is believed that this exchange has now been eliminated and that measurement can be made in the immediate future on TCN.

INFRARED SPECTROSCOPY

Project IR-1 – Infrared and Microwave Spectra of Formyl and Deutero-Formyl Fluorides

P. A. Staats H. W. Morgan
J. H. Goldstein

Work on the infrared spectra of formyl and deutero-formyl fluorides has been completed. An article summarizing the results has been accepted for publication by *The Journal of Chemical Physics*.

No further investigations were made of the microwave spectrum. A joint study with A. H. Nielsen, of the University of Tennessee, of the rotational fine structure of the absorptions in both HCOF and DCOF is being continued, in an attempt to evaluate the rotational and centrifugal distortion constants.

Project IR-2 – Infrared Spectra of Tritium Compounds

P. A. Staats H. W. Morgan
J. H. Goldstein

The spectra of T_2O and THO have been reexamined, with the use of a second preparation of tritium oxide. As in the previous work, about 135 curies was employed. The region of 2- to 6-μ wavelengths was studied with an LiF prism, which gave a resolution of about 1 cm^{-1}.

The fundamentals ν_2 and ν_3 of the HTO molecule have been assigned. In addition to the T_2O fundamentals previously reported,[5] two combination bands have been identified. By exchange, the sample was allowed to pick up a small quantity of deuterium, and one of the TDO fundamental frequencies was observed. Table 1 summarizes the experimentally determined frequencies.

The zero-order fundamental frequencies, computed

[3]Consultant, Emory University, Emory University, Georgia.

[4]Summer research participant, Tennessee Agricultural and Industrial State College, Nashville, Tennessee.

[5]P. A. Staats, H. W. Morgan, and J. H. Goldstein, *Stable Isotope Research and Production Semiann. Prog. Rep. May 20, 1955*, ORNL-1908, p 4.

TABLE 1. CALCULATED AND OBSERVED VALUES OF THE FUNDAMENTALS OF THE ISOTOPIC WATER MOLECULES T_2O, HTO, DTO

Molecule	Fundamental	Calculated Values (cm^{-1})	Observed Values (cm^{-1})
T_2O	ν_1	2235	*a*
	ν_2	996	995.5
	ν_3	2368	2365–2372[b]
	$\nu_2 + \nu_3$	3364	3358[c]
	$\nu_1 + \nu_3$	4603	4537[c]
HTO	ν_1	2291	*a*
	ν_2	1340	1324
	ν_3	3708	3708[c]
DTO	ν_1	2304	Not observed
	ν_2	1092	Not observed
	ν_3	2737	2730[c]

[a] These bands observed, but, due to serious band overlaps, no meaningful center can be reported.

[b] A strong Q branch obscures the exact center of this band.

[c] These values represent the measured centers of the Q branches. Actual band centers appear to be at slightly higher frequencies.

by Libby,[6] have been corrected for the effect of the anharmonicity in the potential function. These corrected values, directly comparable to the observed frequencies, are given for comparison in Table 1. The agreement in all cases is satisfactory.

Project IR-3 – Molecular Force-Field Computations by the Oracle

H. W. Morgan J. H. Goldstein

In conjunction with members of the Mathematics Panel, studies are being made of the computation of F matrix elements, diagonal and off-diagonal, equal in number to the isotopic fundamental frequencies available. As previously reported,[7] a method had been devised by which the Oracle could be employed to determine the diagonal F matrix in the secular equation $FG - \lambda E = 0$. This method was iterative and was time-consuming because the matrix $F^{1/2}GF^{1/2}$ had to be computed for each cycle, since the Oracle could determine only the eigenvalues of symmetric matrices. The time factor thus opposed an iterative approach to the off-diagonal elements, involving at each cycle the above iterative procedure.

In considering this problem, members of the Mathematics Panel have evolved a direct method for the computation of the eigenvalues of nonsymmetric matrices.[8] This procedure, which is now being coded for use, will allow the iterations required for the determination of off-diagonal elements to be performed in a reasonable amount of time.

OPTICAL SPECTROSCOPY

Project OS-1 – Zeeman Spectroscopy

P. M. Griffin K. L. Vander Sluis

During the past six months, methods have been developed which permit operation of electrodeless discharge sources in magnetic fields of 33,500 oersteds, although the magnet gap is only ¼ in. Previously, work in this laboratory was limited to fields of 20,000 oersteds or less. At field strengths of 33,500 oersteds, electrodeless discharge tubes excited by 2450-megacycle radiation and an Echelle-Littrow spectrograph were used to obtain an effective Zeeman resolution of 0.05 Lorentz units; that is, Zeeman components separated by as little as 0.05 Lorentz units can be resolved. An example of the quality of the patterns obtained is given in Fig. 1, which shows the Zeeman patterns of the $^3S_1$-$^3P_{0,1,2}$ triplet of cadmium obtained at a field strength of 33,500 oersteds.

Other instrumental improvements made during the past six months include the revamping of the spectrograph and external optical system to eliminate two reflecting mirrors and one lens outside the spectrograph and the replacing of an aluminized plane mirror with a totally reflecting 45-deg quartz prism inside the instrument; these changes have reduced exposure times required for lines in the ultraviolet region. Both the Echelle and

[6] W. F. Libby, *J. Chem. Phys.* 11, 101 (1943).

[7] P. A. Staats, H. W. Morgan, and J. H. Goldstein, *Stable Isotope Research and Production Semiann. Prog. Rep. May 20, 1955*, ORNL-1908, p 3.

[8] A. C. Downing and A. S. Householder, *Some Inverse Characteristic Value Problems Which Arise in the Study of Simple Molecules*, ORNL CF-55-10-95 (Oct. 20, 1955).

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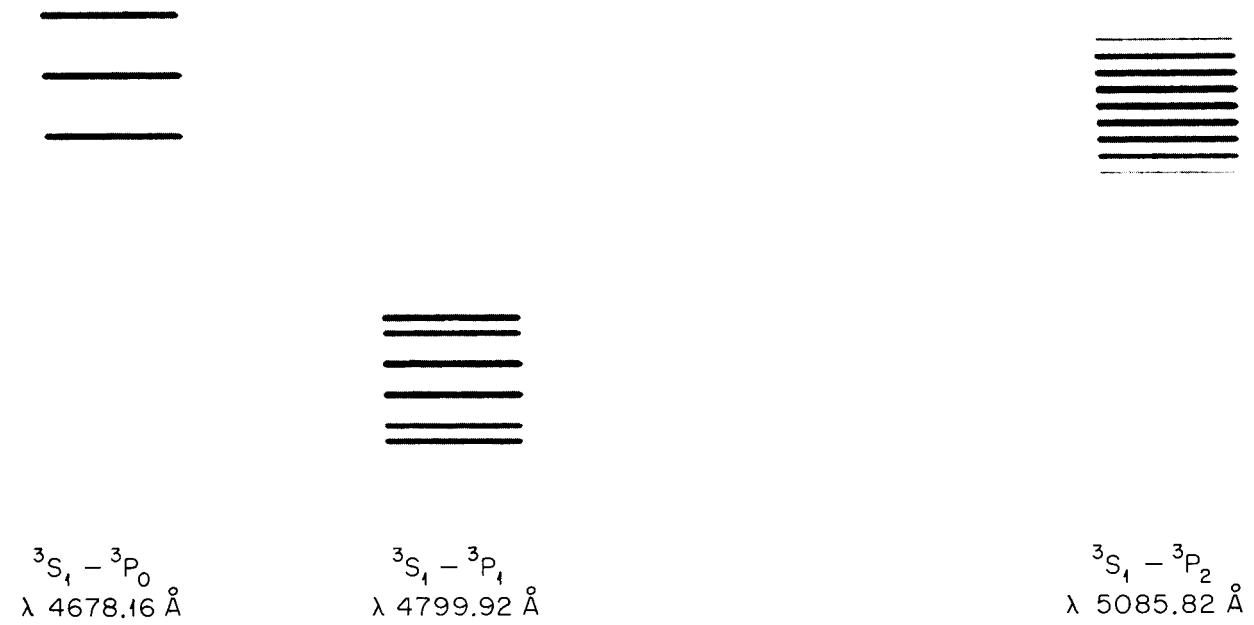

Fig. 1. Zeeman Splitting in the $^3S_1$-$^3P_{0,1,2}$ Triplet of Cadmium at 33,500 Oersteds as Photographed with an Echelle-Littrow Spectrograph.

the Littrow spectrographs have been enclosed in a single box to prevent the lines from broadening because of air currents and air turbulence in the optical paths within the spectrographs. The coils of a sensitive resistance thermometer have been mounted in the box; this arrangement permits continuous monitoring of the temperature during exposures.

To obtain Landé g factors from Zeeman splittings of spectrum lines, it is necessary to know the magnetic field strength in the region occupied by the emitting atoms. This field strength may be measured by some independent method, or it may be calculated from measurements of the splittings of lines from elements present in the source for which the g factors of the levels are known. The latter method has been used by this group for all work to date. Neon is generally used as a support gas in the sources and hence has been convenient for use in field measurements. Since the $1s_5$ level is the only $J = 2$ level arising from the $2p^5(^2P^0_{1\frac{1}{2}})3s$ configuration of Ne(I), the g sum rule requires that its g factor be exactly the same as the g factor for a $^3P_2$ level which is corrected for the anomalous g factor of the electron (see Project OS-3), 1.50115. By measuring lines involving this level or other levels whose g factors are known exactly from the g sum rule, field-strength determinations at 33,415 oersteds have been made to a precision of ±3 oersteds based on a 50% confidence level or to ±10 oersteds on a 95% confidence level.

The Zeeman spectrum of polonium is a problem of considerable importance to this group. However, because of the rather hazardous nature of the element, it seemed advisable to use some other element with similar properties for preliminary work such as sample handling, source design and operation, and investigation of the nature of the spectra obtained. As a result, extensive work has been carried out on tellurium not only because its properties are quite similar to those of polonium but also because information on the tellurium spectrum itself is incomplete.

Zeeman spectra of tellurium, from a sample enriched to 97% Te^{130} to rule out extraneous effects from hyperfine and isotope structures, have been photographed at several densities in the region 2600 to 6500 A. The range will later be extended down to 2200 A and up to 9200 A. Measurement of patterns and calculation of g factors have been made in the region 4600 to 6500 A. From the completed results to date, five new energy levels have been found and their g factors and total angular-momentum quantum numbers J determined. The levels involved in 25 previously unclassified lines have been determined and 112 g factors with their probable errors have been calculated by using the method of least squares described in another section of this report (see Project OS-2). Although the measurements and calculations are not complete, enough has been done to indicate the quality of results to be expected.

The probable error in the g factor for a given level from a single determination is dependent on the total angular-momentum quantum number J as well as on the precision of the measurements. Levels of high J lead to Zeeman patterns of many components which provide more degrees of freedom for the determination. The average probable errors for levels of different J's based on measurements of 43 patterns are illustrated in Table 2.

When more than one line involving a given level is available for the determination of the g factor, a much better value can be obtained by taking a weighted mean of the independent determinations. The weights assigned to each value are proportional to the square of the reciprocal of the probable error for that value, as indicated in the following expressions:

$$\bar{g} = \frac{\sum_i^n (1/r_i^2) g_i}{\sum_i^n (1/r_i^2)},$$

$$r_g = t \sqrt{\frac{\sum_i^n (1/r_i^2)(\bar{g} - g_i)^2}{(n-1)\sum_i^n (1/r_i^2)}}$$

To justify the taking of a weighted mean, it was necessary to demonstrate that the precision of individual determination represented accuracy. The method used was the following: For ten levels which had at least four independent determinations of the level g factor, the average of the individual probable errors,

$$r_b = t \sqrt{\frac{\sum_i \sigma_2^2}{3}},$$

and the average scatter of the individual values about a raw average,

$$r_a = t \sqrt{\frac{\sum_i (\text{residual})_i^2}{3}},$$

were calculated. It was found that, over ten levels, the average of r_a was 0.0025 and that the average of r_b was 0.0023. The close correspondence of the two averages indicates that the precision does represent an accuracy. It should be noted that all these measurements came from a single exposure where the patterns ranged all the way from very weak to very heavy.

The results obtained for ten levels (five of even parity and five of odd parity) are shown in Table 3. For sake of comparison, the g values reported by Mack *et al.*[9] and by Loring and Green[10] are listed in the last two columns.

TABLE 2. AVERAGE PROBABLE ERRORS* FOR A SINGLE DETERMINATION AS A FUNCTION OF THE J OF THE LEVEL

J	Average Probable Errors	Number of Determinations
1/2	0.0030	13
3/2	0.0015	47
5/2	0.0012	23
7/2	0.0011	3

*Based on measurements of 43 patterns in the Zeeman spectrum of Te(II) at 33,500 oersteds.

[9] J. E. Mack *et al.*, *Phys. Rev.* 83, 654 (1951).

[10] J. B. Green and R. A. Loring, *Phys. Rev.* 90, 80 (1953).

TABLE 3. WEIGHTED MEAN g FACTORS FOR ENERGY LEVELS OF SINGLY IONIZED TELLURIUM, Te(II)

Level	Weighted Mean g Factor	Probable Error	Results of Green and Loring	Results of Mack *et al.*
$81_{3/2}$	1.1238	±0.0006	1.08	1.12
$83_{1/2}$	1.0559	±0.0001	1.02	1.07
$86_{3/2}$	0.9353	±0.0009	0.93	0.99
$95_{3/2}$	0.9760	±0.0003		
$78_{1/2}$	2.2954*	±0.0024	2.25	2.4
$99^{\circ}_{3/2}$	1.2572	±0.0007	1.25	1.30
$101^{\circ}_{3/2}$	1.3106	±0.0007	1.30	1.30
$103^{\circ}_{3/2}$	1.7389	±0.0007	1.72	1.74
$105^{\circ}_{5/2}$	1.2898	±0.0003	1.27	1.29
$101^{\circ}_{1/2}$	2.3272*	±0.0014	2.33	2.29

*Unweighted mean.

Project OS-2 – Least-Squares Reduction of Zeeman Data

K. L. Vander Sluis P. M. Griffin
J. H. Vander Sluis[11]

The application of high-resolution source and instrumental techniques to Zeeman studies has resulted in data which potentially can yield Landé g factors of high precision. In order to make full use of this potentiality, it is desirable that the reduction of these data produce the "best values" of the g factors and a measure of precision. Also, considering the volume of data, the reduction should be of a form which lends itself to coding for calculation by a high-speed calculating machine such as the Oracle. The method of reduction which satisfies all three conditions is the method of least squares.

If an atomic energy level of total angular-momentum quantum number J and magnetic moment μ is subjected to a magnetic field H, it will give rise to $2J + 1$ energy levels. The separations of these levels from the original level are given by the expression

$$-\Delta T_k = m_k \cdot g \cdot L \text{ cm}^{-1} ,$$

where

$-\Delta T_k$ = the energy separation in wave numbers;
m_k = the magnetic quantum number having the values $m_k = \pm(J - k)$, where $J \geq k \geq 0$ (k an integer);

$$g = \frac{\mu}{J(J+1)} \frac{4\pi mc}{eh} ;$$

$$L = \frac{He}{4\pi mc^2} .$$

If now the transition $J_2 \rightarrow J_1$ takes place in the magnetic field H, it will produce a Zeeman pattern whose components are transitions between sub-levels satisfying the selection rules $\Delta m = 0$ or ± 1, except for $J_1 = J_2$ and $m_2 = 0$, in which case $\Delta m = \pm 1$. The displacement from zero field position of these components may then be expressed by the set of linear equations

$$r_1 = a'_{11} g_1 + a'_{12} g_2 ,$$
$$\vdots$$
$$r_{n'} = a'_{n'1} g_1 + a'_{n'2} g_2 ,$$

where the set of coefficients a'_{i1}, a'_{i2} is the same set as the magnetic quantum numbers. Determination of the least-squares values of g_1 and g_2 from the set of readings r_i leads to equations in which the

[11] Mathematics Panel.

r_i's occur only as the differences $(r_i - r_{n'})$, $(r_2 - r_{n'-2}) \ldots (r_i - r_{n'-i})$, that is, the differences between symmetric components.

According to Catalan[12] the most common perturbation of the Zeeman pattern (weak Pachen-Back effect) results in shifts of individual components of the Zeeman pattern but in such a manner as to leave unchanged the symmetric differences $y_i = (r_i - r_{n'-i})$. Since these are the quantities occurring in the least-squares solution, the g factors may be expressed as

$$y_1 = a_{11}g_1 + a_{12}g_2 \ ,$$
$$\vdots$$
$$y_n = a_{n1}g_1 + a_{n2}g_2 \ ,$$

where

$$a_{i1} = 2a'_{i1},$$
$$a_{i2} = 2a'_{i2},$$
$$n = n'_2.$$

The least-squares values for the g factors are

$$g_1 = \frac{1}{ac - b^2}(cz_1 + bz_2) \ ,$$

$$g_2 = \frac{1}{ac - b^2}(bz_1 + az_2) \ ,$$

with the variances

$$\sigma^2_{g_1} = \frac{c}{ac - b^2} S^2 \ ,$$

$$\sigma^2_{g_2} = \frac{a}{ac - b^2} S^2 \ .$$

The quantities a, b, c, z_1, z_2, and S^2 have the following values:

$$a = \sum_1^n a_{i1}^2 \ ,$$

$$b = \sum_1^n a_{i1}a_{i2} \ ,$$

$$c = \sum_1^n a_{i2}^2 \ ,$$

$$z_1 = \sum_1^n a_{i1}y_i \ ,$$

$$z_2 = \sum_1^n a_{i2}y_i \ ,$$

$$S^2 = \frac{\sum_1^n (y_{obs} - y_2)^2}{n - 2} \ .$$

An Oracle code has been written so that, if information is given as to the J's of the transition, the positions of the components as read on a coordinate comparator, and the order of interference (m), the machine will calculate the vacuum wave number of each component, the symmetric differences y_i, and the g factors. The printout of the machine consists of the following data: the center of gravity of the Zeeman pattern in vacuum wave numbers, the g factors and their respective probable errors, and the measured y_i's with their respective deviations from calculated y_i's. In addition to printing out this information, the machine will also store these data on magnetic tape in anticipation of future codes which will aid in the spectrum classification.

Project OS-3 – Anomalous Magnetic Moment Correction to Landé g Factors

J. R. McNally, Jr.

In 1933, Pauli[13] pointed out the possibility of a small, intrinsic magnetic moment for the electron. The first experimental evidence from Zeeman data was reported in 1948 by McNally.[14] Koenig, Prodell, and Kusch[15] have reviewed critically the theoretical and experimental evidence for this effect and have reported new evidence. Inasmuch as the evidence for this effect is now overwhelming, the theoretical Landé g factors should be corrected. This correction has been calculated, using the μ_s value of Koenig, Prodell, and Kusch, and the corrected g factor may be obtained by the relation

$$g' = g + 0.00229\ (g - 1) \ ,$$

[12] M. A. Catalan, *J. Research Nat. Bur. Standards* 47, 502 (1951).

[13] W. Pauli, *Quantentheorie*, vol. 24, sec. 1, p 211, in *Handbuch der Physik*, Julius Springer, Berlin, 1933.

[14] J. R. McNally, Jr., *Phys. Rev.* 73, 1130 (1948).

[15] S. H. Koenig, A. G. Prodell, and P. Kusch, *Phys. Rev.* 88, 191 (1952).

where g' is the corrected Landé g factor and g the uncorrected one.

The magnitude of g sums is affected by this correction, but the corrected g sums still exemplify the g sum rule; that is, the corrected g sum is a constant independent of the coupling.

Project OS-4 – Compilation of Data on Thorium Spectra

G. W. Charles

The compilation of information on the spectra of Th(IV) and Th(III) has been completed in rough-draft form. The information includes classified line wavelengths, vacuum wave numbers, deviations between observed and calculated vacuum wave numbers, transitions, and observed Zeeman patterns.

Project OS-5 – Spectrum[16] of Po_2

G. W. Charles

Approximately 200 band heads of Po_2^{210} and 200 heads of Po_2^{208} have been assigned to vibrational transitions involving 57 vibrational levels of the lower state and 31 levels of the upper state. The frequencies have been represented within an average deviation of 0.25 K (K, Kayser = 1 cm^{-1}) by a set of empirical energy levels for each isotope. The energy levels for the lower state of Po_2^{210} are representable within 1.3 K on the average by the equation

$$G(v'') = 0 + 155.78\, v'' - 0.360\, v''^2 \ ,$$

while those for the upper state of Po_2^{210} are representable within 0.4 K by the equation

$$G(v') = 25{,}125.7 + 108.12\, v' - 0.443\, v'^2 \ .$$

New equations calculated from these, by using the theory of the vibrational isotope effect, represent the energy levels of Po_2^{208} within the same limits. Over 100 of the remaining unassigned bands fit well into the present system. Preliminary indication has been obtained of the presence of another system. Consideration of the spectrum is continuing.

Project OS-6 – Ebert Monochromator Studies

G. K. Werner

Critical inspection of the optical quality of each of the reflectors for the Ebert monochromator revealed that the mirror clamps were distorting the spherical mirrors enough to make them astigmatic. When the mirrors were tested unclamped, supported by two pins 3 in. apart, no astigmatism could be detected. When they were reassembled, spherical aberration could still be noticed, especially in low orders, because the unsymmetrical mirror arrangement (6- and 10½-ft focal lengths) does not provide the aberration correction of a conventional Ebert mounting. Another 10½-ft mirror has been obtained in order to make the system symmetrical and thus produce better aberration correction.

At high angles of incidence the focus of the system was found to change as much as 4 cm, the direction of change depending on which side of normal incidence was being tested. Calculations showed that this corresponds to an error of run, the grating constant being about 0.0007% greater at one end than at the other.[17] If this error is gradual and continuous across the grating surface, it will present no difficulties, other than the focal plane shift, in the operation of the spectrograph.

Project OS-7 – Osram Lamp Power Supply

O. B. Rudolph

Standard light sources emitting a light of known wavelength are extremely convenient to have available in any spectroscopic laboratory. Outstanding among such sources are the Osram laboratory lamps,[18] which are available in the following types: sodium, mercury, cadmium, cadmium-mercury, and zinc. One disadvantage of these lamps is the necessity of an a-c supply voltage of 200 v minimum for striking, even though the operating voltage across the lamp is only 15 to 30 v after starting.

A circuit which utilizes a 115-v supply is shown in Fig. 2. In lieu of high line voltage for striking, the inductive kick of the choke is utilized. This choke should have a current-carrying capacity of 3 amp and an inductance of approximately 75 mh (if not obtainable, the choke can be constructed by winding about 50 turns of the proper size wire on an old 1 × 1 in. transformer core). The dpdt relay K_1 is an added refinement, which, though not absolutely necessary, helps to prolong the life

[16] This is a continuation of work initiated at the Mound Laboratory.

[17] Bausch & Lomb grating No. 130 A12 (600 lines per millimeter).

[18] Manufactured by General Electric Company, Ltd., of England.

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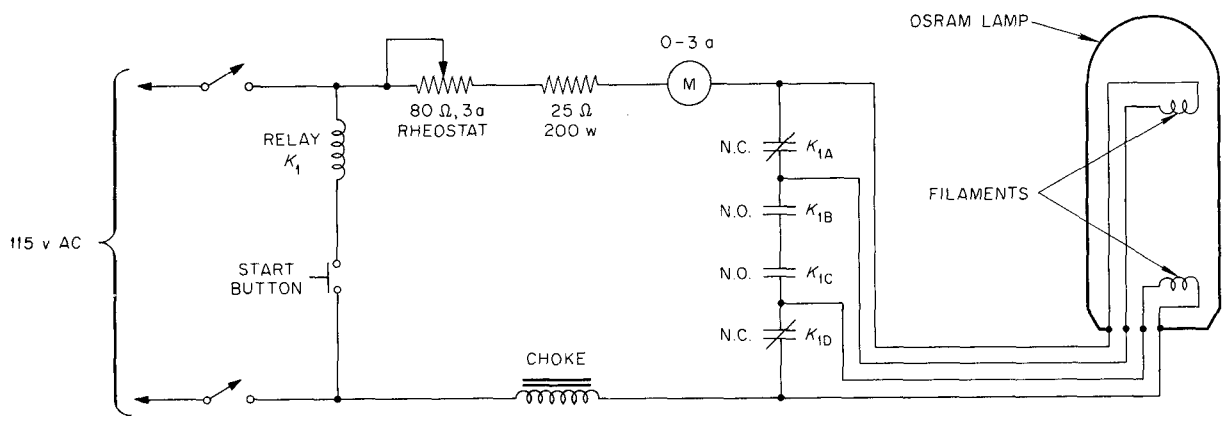

Fig. 2. 115-v Power Supply for Osram Lamps.

of the tube. Without K_1, observation discloses that one end of each filament becomes extremely hot while the other end remains comparatively cool. This is probably due to a slight potential difference from end to end of each filament coupled with any physical difference in the spacing between filaments. By the relay being incorporated as shown, after the tube strikes, both ends of each individual filament are at equal potential and the heat is more evenly distributed.

A 0- to 3-amp a-c meter (M) should be incorporated in the circuit, and care should be exercised that the current rating of the particular lamp used is not exceeded, either in starting or operating. In operation the starting switch should be held closed and the filament current adjusted to the proper value, held for about 10 sec, and then released. It is sometimes necessary to repeat the starting procedure several times in order that the release of the start switch may be in proper phase relation to the a-c voltage peak.

SPECTROCHEMISTRY

Project SC-1 – General Spectrochemistry

Z. Combs M. R. Skidmore
J. A. Norris

A method has been developed for the analysis of certain rare earths in a Y_2O_3 matrix. Individual quantitative standards have been prepared for La (20–200 ppm), Ce (1,000–20,000 ppm), Sm (40–400 ppm), Eu (20–200 ppm), Gd (20–200 ppm), and Dy (100–1000 ppm). A composite standard containing all rare earths, except lutetium, with concentrations varying from 10 to 4000 ppm, depending on the element, has been prepared but, as yet, has not been cross-checked with the individual standards. Numerous special samples of Y_2O_3 have been analyzed, and at present pure Y_2O_3 is being evaluated as a carrier for the determination of less than 1 ppm rare earths in metal samples.[19] Initial results are very favorable.

Project SC-2 – Rare Earths

J. J. Mundzak J. A. Norris

Difficulties have arisen in the use of certain analytical lines because of the changing matrix of samples submitted by the rare-earth separations group. Effort at present is being concentrated on the determination of terbium in dysprosium mixtures. The disagreement in general occurs below the 5% level of Tb in mixtures of Dy, Ho, Er, and Gd. The main problem arises in maintaining the desired sensitivity (1% Tb) in these mixtures.

Project SC-3 – Photoelectric Spectrochemistry

S. J. Ovenshine J. A. Norris

The desired precision for an analytical method of stainless steel analysis with the use of a d-c arc has not been attained. In order to determine

[19] In collaboration with ORNL Y-12 Analytical Chemistry Division.

the reason for the poor precision, an intensive investigation of several factors has been undertaken. In particular, the effects of line reversal, background, and sample homogeneity are being studied. Most instrumental factors have been corrected, and independent check procedures have been established as outside controls. These studies should furnish information as to the validity and usefulness of the method being used.

Project SC-4 – Spectrography of Uranium Isotopes

M. R. Skidmore G. E. Peterson[20]

Sufficient data have been obtained during the last six months on the isotopic analysis of U^{235} in U^{238} to permit statistical treatment of single and quadruplicate determinations. The spectrographic procedure, based on the Jarrell-Ash 3.5-m Ebert spectrograph, was described previously.[21] Results on the determination of U^{235} in a mixture of U^{235} and U^{238} are given in Table 4. The results indicate the advantage of using different line pairs at different concentration levels.

Concern has been expressed about the analysis of U^{235} with small amounts of U^{236} present in the U^{235}-U^{238} mixture. Standards containing 0.50%, 1.00%, 1.48%, and 2.85% of U^{236} in U^{235}-U^{238} mixtures have been made, and preliminary investigations indicate a limit of detection of about 0.5% U^{236}. Quantitative determinations may be made on the 1.48 to 2.85% U^{236} range with a standard deviation of approximately 0.2% U^{236} on duplicate determinations.

[20]On loan from Babcock & Wilcox Co.

[21]M. R. Skidmore, *Stable Isotope Research and Production Semiann. Prog. Rep. May 20, 1955*, ORNL-1908, p 12.

X RAYS

Project X-1 – X-Ray Diffraction

H. W. Dunn

Work is continuing on the tabulation of x-ray diffraction data for the compounds studied in this laboratory for which no x-ray data are available. Data on the interplanar spacings and line intensities will be sent to the American Society for Testing Materials to be published in the next edition of the ASTM data cards. Data for one of these compounds, $UO_3 \cdot 2H_2O$, are included in Table 5. Film was obtained by use of a copper target with a nickel filter.

Project X-2 – X-Ray Spectroscopy

W. F. Peed G. L. Rogosa[22]

Precision wavelength measurements have been made of 4 K lines of technetium, 8 L lines of plutonium, 5 L lines of americium, and 5 L lines

[22]Summer employee, 1955, from Florida State University, Tallahassee, Florida.

TABLE 4. ANALYTICAL ACCURACY OF URANIUM ISOTOPE ASSAY

U^{235} (%)	σ Line Pair 4244.122 (.9F)* / 4244.37 (XF)**		σ Line Pair 4244.122 (.9F) / 4252.426 (XF)	
	Single Analysis (%)	Quadruplicate Analysis (%)	Single Analysis (%)	Quadruplicate Analysis (%)
96.00	0.21	0.14	0.43	0.28
93.09	0.37	0.20	0.50	0.31
88.55	0.54	0.32	0.75	0.34
84.13	0.60	0.31	0.68	0.35
80.02	0.96	0.51	0.95	0.52
75.67	1.23	0.86	1.02	0.57

*.9F signifies 0.9 density filter used.

**XF signifies no filter.

TABLE 5. X-RAY DIFFRACTION DATA ON $UO_3 \cdot 2H_2O$

Intensity	Spacing, *d* (A)	Intensity	Spacing, *d* (A)
Cut off	23.28	20	1.629
40	7.47	15	1.609
100	7.24	5	1.575
3	4.43	10	1.561
2	4.04	3	1.533
2	3.75	2	1.515
60	3.65	1	1.480
60	3.57	5	1.445
60	3.47	2	1.419
3	3.38	5	1.358
5	3.30	2	1.336
70	3.20	2	1.321
55	3.15	3	1.303
2	3.05	1	1.292
3	2.99	2	1.283
2	2.95	1	1.257
2	2.92	2	1.228
10	2.87	2	1.215
5	2.83	2	1.204
5	2.79	7	1.189
2	2.75	1	1.177
3	2.60	1	1.171
40	2.55	2	1.163
20	2.52	3	1.141
3	2.48	3	1.132
10	2.44	2	1.124
2	2.38	2	1.113
10	2.30	2	1.041
5	2.26	3	0.998
3	2.21	3	0.981
1	2.18	2	0.962
10	2.14	2	0.951
2	2.11	1	0.945
7	2.06	1	0.940
20	2.03	2	0.933
30	2.01	1	0.927
50	1.954	2	0.923
3	1.909	2	0.911
2	1.851	1	0.902
10	1.829	2	0.891
10	1.808	2	0.879
50	1.776	2	0.869
15	1.745	1	0.857
1	1.721	1	0.843
7	1.699	1	0.813
5	1.681	Edge	0.773

of neptunium. Significant differences between the results obtained by workers in this laboratory and those by other workers were observed for the transuranic elements. Agreement with measurements of Cauchois *et al.*[23] for plutonium was found. Results for neptunium, which are typical of the neptunium, plutonium, and americium results, are shown in Table 6. A detailed report of this work is scheduled for publication in *The Physical Review* in January 1956.

[23]Y. Cauchois *et al.*, *Compt. rend.* 25, 1782 (1955).

Project X-3 – X-Ray Fluorescence Studies

W. F. Peed

Application of methods of x-ray fluorescence to analysis of uranium alloys is being reinvestigated. Alloy systems being considered include Zr-U, Nb-U, Mo-U, and Zr-Nb-U. Preliminary results show that analysis of the metal can be made directly – that is, without chemical treatment – in 15 min. The precision compares favorably with that of chemical methods.

TABLE 6. WAVELENGTH MEASUREMENTS OF NEPTUNIUM L X RAYS

Line	ORNL Data (kev)	Data of Jaffe *et al.*[a] (kev)	Data of Day[b] (kev)	Extrapolated Values of Hill *et al.*[c] (kev)
α_2	13.758 ± 0.004	13.79 ± 0.04	13.776 ± 0.003	13.76
α_1	13.960 ± 0.003	13.98 ± 0.04	13.961 ± 0.003	13.95
β_2	16.883 ± 0.005	16.89 ± 0.02	16.857 ± 0.006	16.84
β_1	17.745 ± 0.005	17.78 ± 0.01	17.764 ± 0.003	17.74
γ_1	20.781 ± 0.007	20.82 ± 0.02	20.796 ± 0.005	20.77

[a]H. Jaffe *et al.*, *Phys. Rev.* 97, 142 (1955).
[b]P. Day, *Phys. Rev.* 97, 689 (1955).
[c]R. D. Hill, E. L. Church, and J. W. Mihelich, *Rev. Sci. Instr.* 23, 523 (1952).

MASS SPECTROMETER LABORATORY

Russell Baldock, Department Head

INTRODUCTION

Emphasis is continuing on the research and development of high-precision methods of analysis, and, in addition, the program of research has been extended to include some of the more fundamental aspects of atomic and molecular structure. Outstanding achievements during this period are as follows:

1. The mass spectrum of copper iodide has given evidence of vapor polymerization up to Cu_3I_3.
2. The specie Ag_3^+ has been found in the fragments appearing in the mass spectra of silver chloride, silver bromide, and silver iodide.
3. A method has been devised for determining the heat of polymerization.
4. In the mass spectrum of propane, a metastable ion has been found at mass 17.8, as was predicted from theory.
5. The uranium content of the Missouri and Mississippi River waters has been measured by using the isotope-dilution technique.
6. New and simple techniques have been devised for preparing gaseous samples for the isotopic analysis of carbon silicon, sulfur, boron, oxygen, and tritium.
7. The mass spectrometer MS-A has been renovated and restored to operation.
8. The Micro-Chemistry Laboratory has been completed and is occupied.
9. The mass spectrometer MS-D has been equipped with a ratio detector which permits the simultaneous detection of two positive-ion beams.

VAPOR STATE OF INORGANIC SOLIDS

H. M. Rosenstock L. K. Brice, Jr.[1]
J. R. Walton

A number of mass spectra of solids were determined as a continuation of earlier studies.

The mass spectrum of CuI gave evidence for vapor-phase polymerization up to Cu_3I_3. The combined results of studies on the cuprous halides are to be published in *The Journal of Chemical Physics*.

The results obtained with the cuprous halides suggested that a study of the silver halides might be of interest. The mass spectra of AgCl, AgBr, and AgI were obtained, and all three substances showed monomeric and trimeric molecule-ions. No dimeric ions were detected. Among the fragments appearing in the mass spectra were Ag_2 and some indications of Ag_3^+. These are of particular interest in connection with the reported polymerization of metallic silver vapor.[2,3] The mass spectra of the compounds are shown in Table 7.

TABLE 7. APPROXIMATE RELATIVE ABUNDANCES OF IONS FOUND IN MASS SPECTRA OF COPPER AND SILVER HALIDES

Compound	Cu			Ag		
	Cl	Br	I	Cl	Br	I
M	30	10	30	100	20	100
MX	5	15	30	90	30	95
M_2	25	10	30	25	10	25
M_2X	60	50	50	70	45	90
M_2X_2	15	10	7			
M_2X_3		1				
M_3	2	1	5	5	4	
M_3X	2	2		2	3	
M_3X_2	40	65	90	85	100	80
M_3X_3	100	100	100	42	37	60
M_3X_4	1					
M_4						
M_4X						
M_4X_2	1					
M_4X_3	2	2				
M_4X_4	15	1				

Further studies on silver chloride were carried out with the objective of obtaining some information about the energetics of the polymerization reaction

$$3AgCl \rightleftharpoons Ag_3Cl_3$$

[1]Summer research participant, Chemistry Department, Virginia Polytechnic Institute.

[2]A. W. Searcy, R. D. Freeman, and M. C. Michel, *J. Am. Chem. Soc.* **76**, 4050 (1954).

[3]W. A. Chupka and M. G. Inghram, *J. Phys. Chem.* **59**, 100 (1955).

In this connection an approximation method was devised for determining the ratio of the heat of vaporization of one vapor species to that of another. This ratio, when combined with other thermochemical data, was sufficient for the heat of polymerization to be determined.

The silver chloride was vaporized from a small platinum-filament oven in a solids mass spectrometer and ionized by electron impact, and then the $AgCl^+$ and $Ag_3Cl_3{}^+$ intensities were measured at various values of the heating current. Here the study is not of a quasi-equilibrium effusion process but rather of a rate of vaporization. In this paper it is assumed that the rate of vaporization follows an equation of the form

$$\gamma = c \cdot \exp(-\Delta H/RT) \ ,$$

where ΔH is the heat of activation for vaporization, and the ion intensity, P, in the mass spectrometer is assumed to be proportional to the rate of vaporization. Then if the intensity of an ion representative of, say, the monomer at two temperatures is compared with that of a representative of the trimer at the same two temperatures, the simple result

$$\frac{P_1{}^{I}/P_2{}^{I}}{P_1{}^{III}/P_2{}^{III}} = \frac{\Delta H^{\pm I}}{\Delta H^{\pm III}}$$

is obtained, where $P_1{}^{I}$, $P_2{}^{I}$ and $P_1{}^{III}$, $P_2{}^{III}$ are the intensities of the monomer and trimer representatives at temperatures T_1 and T_2, respectively. This leads to the useful premise that a log-log plot of monomer representative vs trimer representative should give a straight line whose slope is equal to the ratios of the heats of activation for vaporization of the two species. It is unnecessary to measure the temperatures.

The results obtained on one experiment with silver chloride are shown in Fig. 3. It is seen that the plot consists of two straight lines. The lower line refers to solid silver chloride, and the higher to the liquid. The experiments were repeated five times and the slopes of the two lines determined by least-square methods. The data for the five experiments are given in Table 8. If it is now assumed that the heats of activation for vaporization and the heat of vaporization are equal,

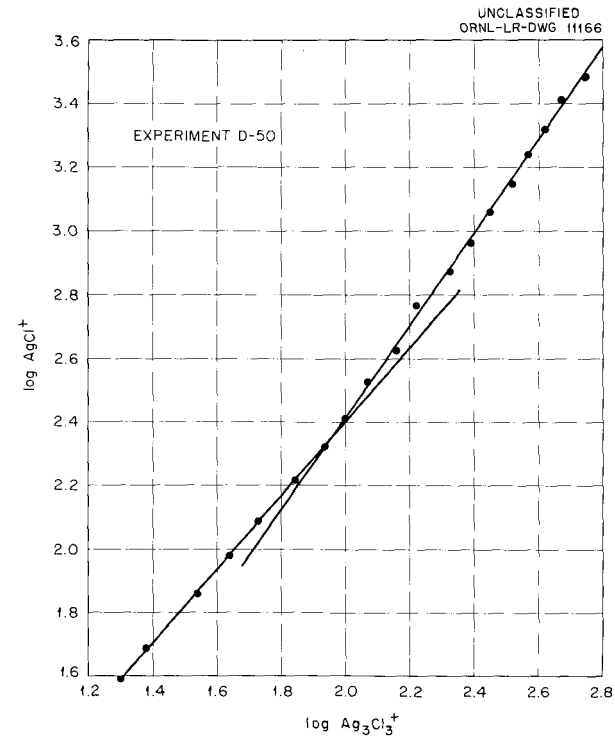

Fig. 3. Log $Ag_3Cl_3{}^+$ vs Log $AgCl^+$ Peak Height.

TABLE 8. HEAT OF SUBLIMATION AND VAPORIZATION RATIOS OF AgCl MONOMER AND TRIMER

Experiment Number	$\frac{\Delta H \text{ Sublimation (Monomer)}}{\Delta H \text{ Sublimation (Trimer)}}$	$\frac{\Delta H \text{ Vaporization (Monomer)}}{\Delta H \text{ Vaporization (Trimer)}}$
D-50	1.16	1.44
D-53	1.19	1.36
C-300	1.20	1.40
D-52	1.26	1.45
D-49	1.23	
Average	1.21 ± 0.03	1.41 ± 0.03

the two ratios can be applied to a calculation of the heat of polymerization. The published value[4] for the heat of fusion of silver chloride is approximately 3.2 kcal/mole, and the value[5] for the heat of vaporization from the liquid state, just above the melting point, is 48 kcal/mole. The latter quantity was determined by vapor-pressure measurements at temperatures considerably above the melting point. The mass-spectrometer data indicate that the vapor at that temperature is probably largely monomeric, and therefore this heat may be taken to be the heat of vaporization of the monomer. Considering now the heat of vaporization ratios and the heat of sublimation ratios determined in the present experiments, it is readily seen that they are consistent with the values quoted above. A heat of polymerization for the reaction

$$3AgCl \rightleftharpoons Ag_3Cl_3$$

can now be calculated to be about 110 kcal/mole at the melting point. Similar studies are being carried out with KCl.

The mass spectra of the following compounds were briefly examined: $FeCl_2$, $PbCl_2$, $NiCl_2$, and SeO_2. No extensive polymerization was found.

METASTABLE TRANSITIONS IN MASS SPECTRA

H. M. Rosenstock C. E. Melton

Some studies have been carried out on metastable transitions in a number of molecules. By using the absolute reaction rate theory of mass spectra,[6] it was predicted[7] that in the propane mass spectrum a metastable transition would occur at mass 17.8 corresponding to the spontaneous process

$$C_3H_8^+ \longrightarrow C_2H_4^+ + CH_4$$

This transition has now been found experimentally.

According to the theory, the parent molecule ion can have only metastable transitions for the decomposition reactions which have the lowest activation energies. The studies of Kambara[8] on propyl alcohol appearance potentials lead, when incorporated with other data, to an activation energy of 3.7 ev for the process

$$C_3H_7OH^+ \longrightarrow C_3H_6^+ + H_2O$$

whereas there are two competing processes which have essentially zero activation energy. Nevertheless, this reaction has a large metastable transition. A calculation of the heat of reaction for this process by using thermal data and ionization potentials indicates the process to be slightly exothermic, a result in agreement with the theory. The discrepancy in Kambara's results, which are too high by about 4 ev, comparable to the value (5 ev) for the OH-bond energy in water,[9] suggests that he measured the appearance potential for the onset of the process

$$C_3H_7OH^+ \longrightarrow C_3H_6^+ + H + OH$$

A cornerstone of the theory of mass spectra is the assumption that there is completely random energy transfer within the molecule-ion by the mechanism of radiationless transitions among electronic states. An example of the limitation of the theory has been found in benzene, where there are four metastable transitions of the parent ion, whose activation energies are 4.7, 5.2, 5.7, and 6.3 ev. In normal hexane, on the other hand, there are five decompositions of the parent ion, whose activation energies are approximately 0 ± 0.4, 0.1, 0.3, 0.3, and 0.7 ev. The only metastable transition found occurs for the 0.1-ev reaction. This indicates that there is incomplete randomization of excitation energy in the benzene, which can be accounted for by the higher symmetry of the molecule, which leads to a bonded structure of electronic states with considerably less crossing of states than in the case of hexane. This is consistent with the nature of the ultraviolet spectra of the two compounds.

MEMORY EFFECTS OF BORON IN THE MASS SPECTROMETER

C. E. Melton L. O. Gilpatrick
Russell Baldock

In earlier studies of the memory effects of boron trifluoride in the mass spectrometer, there was speculation as to the effects of stopcock grease and the extent of isotopic exchange between boron

[4] L. Brewer, paper 7 in *Chemistry and Metallurgy of Miscellaneous Materials: Thermodynamics* (ed. by L. L. Quill), NNES IV-19B, McGraw-Hill, New York, 1950.

[5] K. K. Kelley, *U. S. Bur. Mines, Bull.* 383 (1935).

[6] H. M. Rosenstock *et al.*, *Proc. Nat. Acad. Sci. U. S.* **38**, 667 (1952).

[7] A. Kropf, PhD Thesis, University of Utah (1955).

[8] T. Kambara, *J. Phys. Soc. Japan* **5**, 31 (1950).

[9] T. L. Cottrell, *Strength of Chemical Bonds*, Academic Press, New York, 1954.

in boron trifluoride and boron in pyrex glass.[10] A soft-glass sample reservoir which had been exposed to boron trifluoride was flamed under vacuum for 30 min to remove adsorbed BF_3. The rate of desorption was studied by exposing the surfaces to BCl_3.[11] A control reservoir had 63% greater desorption of BF_3 than did the reservoir which had been flamed, thus indicating that most of the adsorption takes place on the glass rather than in the stopcock grease.

Two 250-cc sample reservoirs, identical except for one being pyrex and the other being soft glass, were filled with BF_3 containing 95.96% B^{10} at a pressure of 2 cm Hg.

Isotopic abundance measurements completed after 128 hr gave a decrease of 2.56% in the abundance of B^{10} in the pyrex reservoir and no detectable change in the abundance of B^{10} in the soft-glass reservoir. These data suggest that there is isotopic exchange between boron in several molecular layers of glass. The detailed processes of the adsorption and desorption of BF_3 are quite complicated, and further studies are now under way. More detailed information may be found elsewhere.[12]

URANIUM IN SEA WATER

L. O. Gilpatrick E. Rona[13]

The analysis of sea water for its uranium content by the isotopic-dilution method was begun in July 1954 and is being continued. A number of results available at that date were given in the previous semiannual report.[14] Additional determinations have been made since then on deep waters of the Gulf of Mexico and on the Missouri and Mississippi Rivers. The bulk of the background uranium reported previously as blank values has been traced to the ion-exchange resin (Dowex-1, 150–200 mesh). Additional samples are still being received for analysis. The values are shown in Table 9.

[10] C. E. Melton, L. O. Gilpatrick, and Russell Baldock, *Stable Isotope Research and Production Semiann. Prog. Rep. May 20, 1955*, ORNL-1908, p 23.

[11] *Ibid.*, p 24.

[12] C. E. Melton *et al.*, *Improved Techniques for the Isotopic Analysis of Boron on the Mass Spectrometer*, ORNL-2006 (in press).

[13] Oak Ridge Institute of Nuclear Studies, joint project.

[14] L. O. Gilpatrick and E. Rona, *Stable Isotope Research and Production Semiann. Prog. Rep. May 20, 1955*, ORNL-1908, p 16.

TABLE 9. URANIUM CONTENT OF SEA AND RIVER WATERS BY ISOTOPIC DILUTION

Body of Water Sampled	Latitude	Longitude	Uranium Content* (10^{-6} g/liter)	Average Uranium Content (10^{-6} g/liter)
Gulf of Mexico				
1800 meters	21°38′N	85°47′W	3.34	
1800 meters	21°38′N	85°47′W	3.33	3.34
2000 meters	23°51′N	86°03′W	2.71	2.71
2000 meters	25°49′N	79°55′W	2.93	2.93
600 meters	27°53′N	86°35′W	3.17	
			3.13	3.15
Mississippi River at St. Louis			1.80	
			1.80	1.80
Missouri River at St. Louis				
Acidified			2.29	
Raw			2.41	2.35
Blanks				
Standard procedure without			0.024	
ion-exchange concentration			0.023	0.024
Standard procedure			0.17	
			0.14	0.15

*All values are corrected for blank contribution.

SPECIAL TECHNIQUES FOR GAS ANALYSIS

C. E. Melton L. O. Gilpatrick

Many isotopic abundance measurements on the mass spectrometer can be completed by using a solid as the sample and performing simple "mass-spectrometry chemistry" to convert to the gaseous state. This mass-spectrometer chemistry consists in heating a solid sample in a quartz container attached to the mass spectrometer. Many solid samples decompose readily into gases upon being heated, and others can be converted by being heated in the presence of other compounds. Some advantages of converting a solid to a gas on the machine are ease in handling the sample, permanent storage of a portion of the sample for future reference, and completion of conversion on a small scale. Some solids which can readily be converted to a gaseous form on the machine are carbon, sulfur, silicon, boron, oxygen compounds, and tritium compounds.

Carbon

Some carbon was converted to carbon dioxide, CO_2, on the G-E analytical mass spectrometer and analyzed for isotopic abundance. This analysis was difficult because most oxygen, O_2, contains CO_2 as a contaminant. Results from preliminary investigations indicated that O_2 obtained from the decomposition of HgO was relatively pure; however, it was necessary to decompose about 5 mg of HgO to flush the vacuum system before O_2 pure enough for the conversion could be obtained. The pure O_2 and carbon were heated to approximately 900°C, and the resultant CO_2 was used for the analysis. This method of conversion is unique in that the O_2 can be monitored for carbon contaminants at the time of the conversion.

Sulfur

Cadmium sulfide, CdS, and sublimed sulfur have been converted to sulfur dioxide in a manner similar to that described for carbon dioxide. The isotopic abundance measurements were made on both the SO^+ and the SO_2^+ ion beams, with a correction being made, of course, for the contribution from the oxygen isotopes. Since the isotopic abundance of the O_2 can be accurately determined on the machine prior to the conversion, the correction factors for the O_2 are well known; hence, an accurate isotopic abundance for the sulfur is obtained.

Silicon

Barium fluosilicate, $BaSiF_6$, has been used for silicon analysis. This compound decomposes at about 400°C, releasing silicon tetrafluoride, SiF_4. Isotopic determinations for silicon were made by using the SiF_3^+ ion beam.

Boron

The calcium fluoride–boron trifluoride complex, $CaF_2 \cdot BF_3$, is readily decomposed by heat into BF_3 for isotopic analysis of boron.

Oxygen

Sodium nitrate, $NaNO_3$, sodium carbonate, Na_2CO_3, and calcium carbonate, $CaCO_3$, have been decomposed and the resultant gases used for isotopic abundance measurements on oxygen. The $NaNO_3$ yields O_2, while Na_2CO_3 and $CaCO_3$ give CO_2.

Tritium

Uranium tritide, UT_3, has been decomposed for isotopic abundance measurements for tritium. A metal vessel containing UT_3 was attached to the mass spectrometer and heated, and then the resultant tritium was assayed. Normally, any excess sample in the mass spectrometer is pumped out after an analysis, but the tritium was reabsorbed in the uranium and only the amount that escaped through the pinhole leak was lost. This reabsorption is unique in that an extremely small portion of the sample was used for the analysis.

STABLE ISOTOPE ANALYSES

J. R. Walton

Seventy-four calutron-enriched isotope samples, received from the Stable Isotope Separation Research and Calutron Operations Department, were analyzed in mass spectrometer MS-C. These samples were composed of 20 different elements. The elements and compounds used and the ions measured during the analyses are shown in Table 10.

Rapid solid analyses techniques, previously described,[15] were successfully employed in the analyses of 6 of the 74 samples.

[15] H. M. Rosenstock, J. R. Sites, and J. R. Walton, *Stable Isotope Research and Production Semiann. Prog. Rep. May 20, 1954*, ORNL-1732, p 22.

TABLE 10. IONS MEASURED IN STABLE ISOTOPE ANALYSES

Atomic No.	Element Investigated	Form Used	Oven	Ions Measured	
				Thermal	Electron-Produced
3	Lithium	Li_2SO_4	Ta	Li^+	
5	Boron	H_3BO_3	Ta		B^+
12	Magnesium	MgO	Ta		Mg^+
19	Potassium	KCl	Ir	K^+	
20	Calcium	CaI_2	Ta		CaI^+
24	Chromium	Cr_2O_3	Ta	Cr^+	
26	Iron	Fe_2O_3, FeI_2	Ir		Fe^+
28	Nickel	NiO	Ta		Ni^+
29	Copper	CuO	Ta		Cu^+
30	Zinc	ZnO	Ta		Zn^+
32	Germanium	GeO_2	Ta		Ge^+
34	Selenium	Se	Ta		Se^+
37	Rubidium	RbCl	Ta	Rb^+	
44	Ruthenium	Ru	Ir	Ru^+	
47	Silver	Ag	Ta		Ag^+
52	Tellurium	Te	Ta		Te^+
57	Lanthanum	La_2O_3	Ta	LaO^+	
60	Neodymium	Nd_2O_3	Ta	NdO^+	
73	Tantalum	Ta_2O_5	Ir		TaO_2^+
78	Platinum	Pt	Ta		Pt^+

TWO-STAGE MASS SPECTROMETER PROGRAM

G. F. Wells J. R. Sites

Renovation of Mass Spectrometer MS-A. – The construction of the new vacuum system and analyzer tube for MS-A has been completed (see Fig. 4) and ion-beam-focusing studies are now being completed.

Several techniques new to this laboratory have been successfully tested on MS-A to check the feasibility of incorporating them in the design of the two-stage, 12-in. machine.[16] Some of the components and results of tests are indicated in the following:

1. The Martin[17] triple-stage mercury diffusion pumps hold the vacuum system at an indicated pressure of 5×10^{-8} mm Hg, as is specified for them in the manufacturer's literature.

2. All joints are Heliarc-welded, and only two vacuum leaks have been found, one of which was not due to improper welding.

3. The Veeco[18] valves and the gold wire gaskets are very satisfactory and can be used easily.

4. The tantalum-faced all-metal valve between the source and the analyzer tube is not completely tight when there is atmospheric pressure on one side and 5×10^{-8} mm Hg pressure on the other. However, after room air leaked through the valve for 2 hr, the tube pressure was raised to 2×10^{-6} mm Hg, and then the source side of the tantalum

[16] G. F. Wells and J. R. Sites, *Stable Isotope Research and Production Semiann. Prog. Rep. May 20, 1955*, ORNL-1908, p 26.

[17] H. S. Martin & Co., 1916–1920 Greenleaf Street, Evanston, Ill.

[18] Vacuum-Electronic Engineering Co., 86-A Denton Ave., New Hyde Park, Long Island, N.Y.

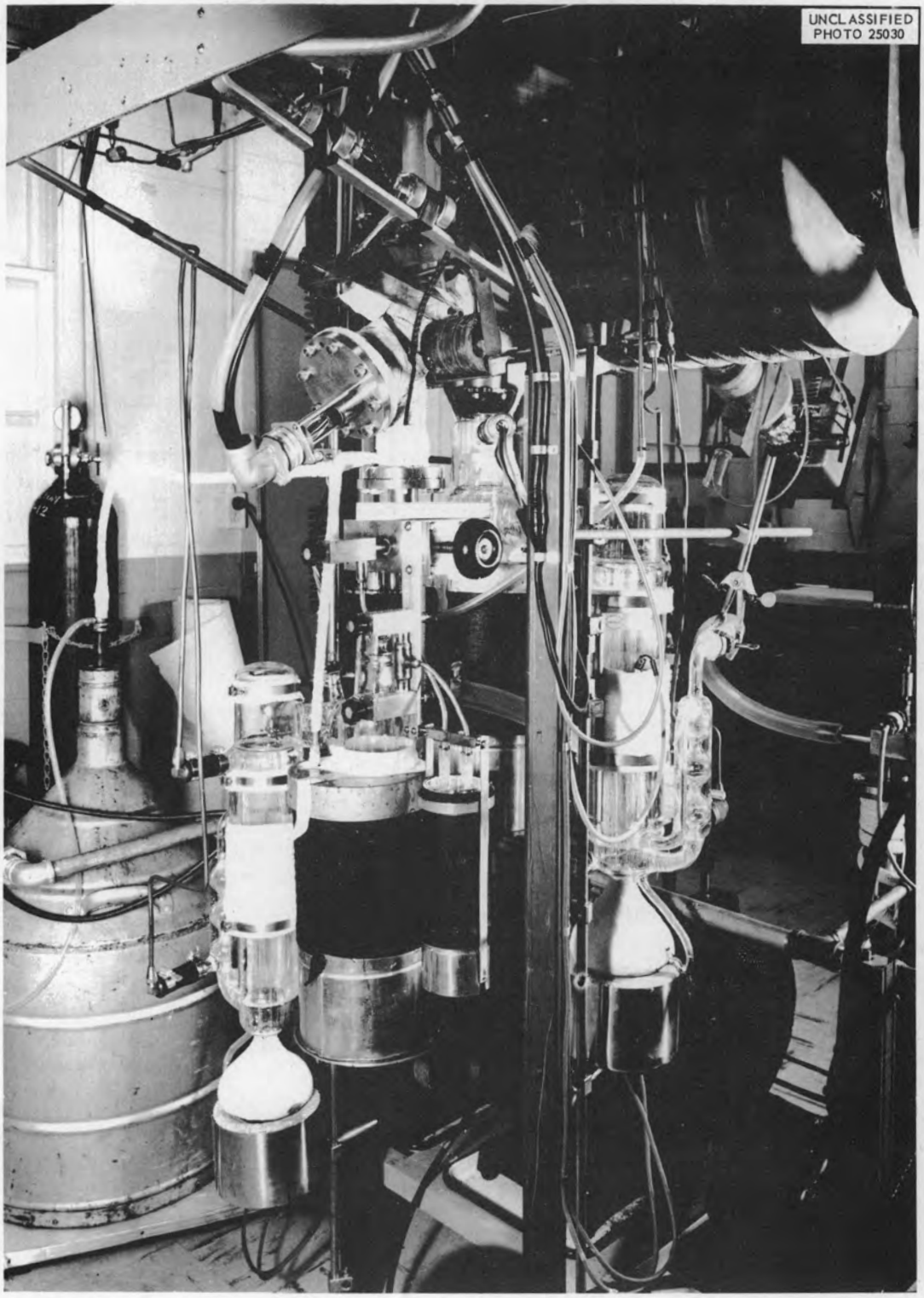

Fig. 4. Source View of MS-A.

valve was pumped down to 2 μ with a fore pump, dropping the tube pressure to 7×10^{-8} mm Hg in 3 min. Thus, this leaking of the tantalum valve is not at all significant.

5. When the Inconel tube was being baked out at 300°C, the distance between punch marks on the source and the receiver was increased, from its value at room temperature, only 0.05%.

6. The Kovar seals are very sturdy, vacuum-tight, and easy to Heliarc weld in position. The receiver-slit rotation and the width-variation mechanism, which uses a stainless steel bellows, both work as designed. As yet, only the usual vibrating-reed ion-detector system has been used. The electronics for the electron-multiplier ion-indicating system is 90% completed.

The Two-Stage Mass Spectrometer MS-F. – The two main magnets for this instrument are illustrated in Fig. 5. They have been designed so that either magnet can be rotated 90 deg for S orientation or U orientation of the tubes. The original design concept called for only the U orientation, and all problems originally postulated[16] can be satisfactorily done with it. However, there is an area of mass spectrometry, as indicated by Inghram and Hayden,[19] in which the S orientation is preferred. By centering the magnets on the bed plate and allowing for 90-deg rotational symmetry of the magnet while the necessary lateral movement is being maintained for ion optical-alignment purposes, the added flexibility of obtaining either tube orientation can be achieved.

Difficulty in procuring certain materials (the yoke and pole iron) has delayed completion of the magnet construction.

The main tubes, source, receiver, and intermediate collector sections are now in the design phase. The Inconel all-metal Heliarc-welded construction techniques are being used. High-temperature bake-out procedures will be used to obtain ultimate base pressures. Application of these techniques in the redesign of MS-A has been beneficial in lowering its base pressure to 5×10^{-8} mm Hg. Certain features of the MS-A design, such as introducing unsymmetrical welds on side-arm tubes, which leads to unaccountable warpage of the main tube, are to be avoided. In general, the quality of the Heliarc welding on Inconel and on Inconel–stainless steel joints is excellent. The all-metal source isolation valve and receiver-adjustment mechanism have proved to be quite satisfactory and are being incorporated in the MS-F design.

The electron multiplier to be used as the primary ion detector is shown in Fig. 6. It is a 14-stage Ag-Mg multiplier with Farnsworth design dynodes. This unit was designed by the Dumont Company[20] to have a gain of 10^7. Glass-sealed 1-megohm Victoreen[21] resistors are to be installed between dynodes to furnish necessary dynode voltages.

The Micro-Chemistry Laboratory. – The auxiliary Micro-Chemistry Laboratory has been completed and is now being occupied. Special attention has been paid to achieving clean operations in this area. All air entering the room is forced through a CWS type of filter, which removes particles down to a few microns in diameter. The room is sealed except for the hood exhaust, which is balanced with the input air to maintain a slight positive pressure. Thus, when either door is opened, air flow is out of the area.

EXPERIMENTAL MASS SPECTROMETER MS-E

G. F. Wells

Design on a mass-spectrometer framework for experimental uses of the instrument was begun during this period. The electrical power installation and associated cabinetry and the tube and magnet framework have been completed. The first proposed instrument to utilize this framework is to be a 6-in. 60-deg-sector tube fabricated in the K-25 mass-spectrometer shops. Its source is designed to enable study of appearance potentials by using an electron multiplier receiver. Appropriate source-gating circuitry, patterned on the one described by Fox *et al.*.[22] has already been developed. Also available is a standard G-E 6-in. 60-deg-sector mass-spectrometer magnet.

The framework, as shown in Fig. 7, is adaptable for other sizes of tubes and magnets without major modifications. Thus, a degree of flexibility is achieved in anticipation of future needs in single-stage mass-spectrometer development.

[19] M. G. Inghram and R. J. Hayden, *Mass Spectroscopy*, p 24, National Research Council, Washington, 1954.

[20] Allen B. Du Mont Laboratories, Inc., 750 Bloomfield Ave., Clifton, N.J.

[21] Victoreen Instrument Co., 3804 Perkins Ave., Cleveland, Ohio.

[22] R. E. Fox *et al.*, *Phys. Rev.* **84**, 859 (1951).

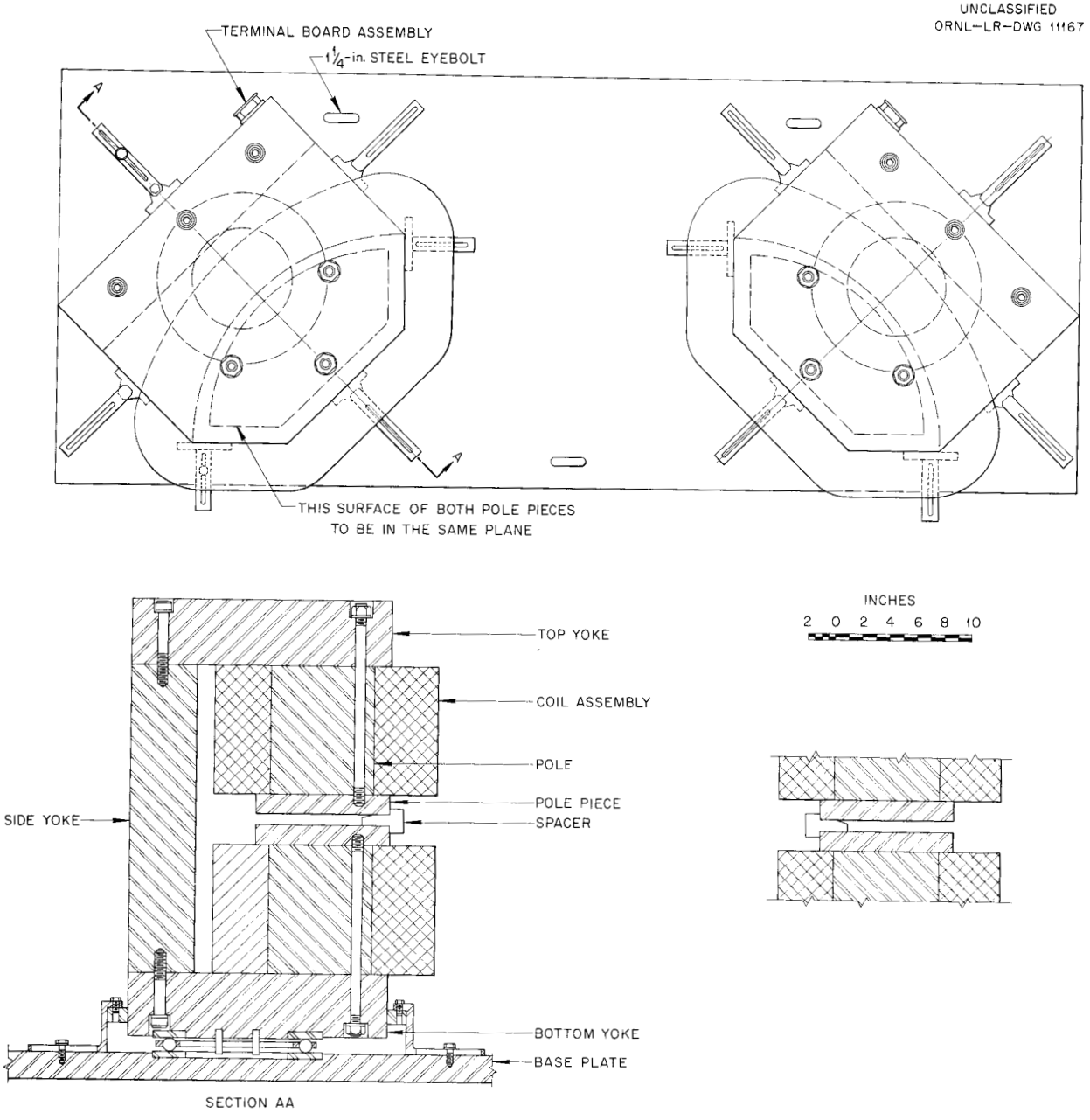

Fig. 5. Two-Stage Mass-Spectrometer Magnets and Bed Plate.

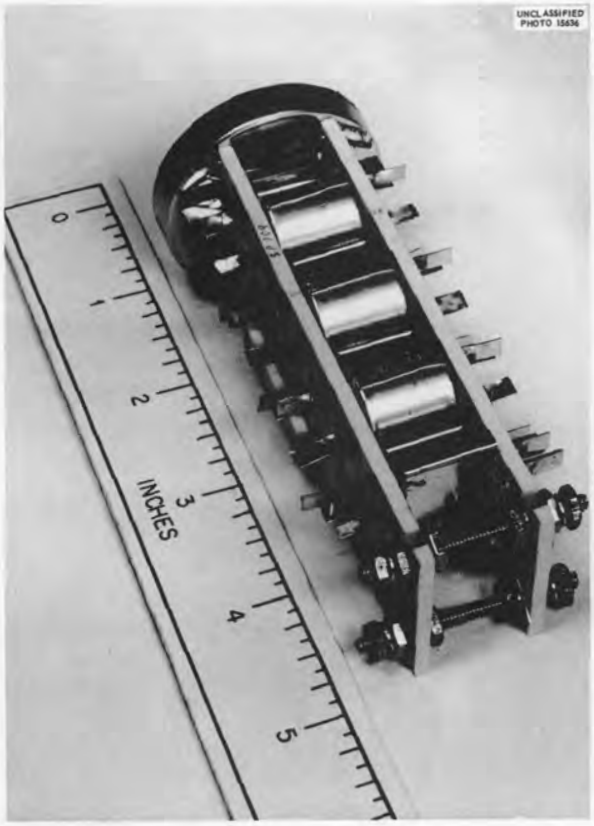

Fig. 6. Electron Multiplier Assembly.

DIRECT DETERMINATION OF ISOTOPE RATIOS

W. R. Rathkamp

A two-ion-beam-collector ratio receiver was designed and installed in MS-D in an attempt to meet the continually increasing demand for greater precision of isotope ratio determinations.

This collector contains two entrance-defining slits in a grounded plate, each wide enough to pass all the beam approaching the receiver. The slits are spaced the correct distance apart to pass simultaneously the two beams of particular interest. Behind these defining slits is placed a second plate with much larger slits, through which the beam passes unhindered. When biased 90 v negative with respect to ground, this plate serves to suppress secondary electron emission from both the defining slits and collector plates. Finally, the beams are received on two flat collector plates placed directly in line with the first defining slits.

Leads from each of these collector plates were brought out of the spectrometer envelope through glass seals and were connected to two vibrating-reed electrometers. The output signal from one reed was passed through a Helipot, and the tapped fraction was bucked against the total output of the other reed, with a center zero recorder used as a null indicator. A simple reading of the Helipot dial gives the ratio of one beam to the other.

In a preliminary test of this system, using potassium as a sample, the K^{39}-to-K^{41} ratio was determined as 13.45, with variations of two units in the last decimal place being readily detected.

While some work still needs to be done in calibration and stabilization of the electronics associated with this method of ratio measurement, the system seems to be capable of delivering determinations of isotope ratios accurate to one part in a thousand.

Fig. 7. MS-E Mass-Spectrometer Frame.

STABLE ISOTOPE SEPARATION AND CALUTRON OPERATIONS

L. O. Love, Department Head

INTRODUCTION

Activities of the Stable Isotope Separation program in the last six months have been directed toward the attainment of certain very high-purity isotopes and replenishment of material in the inventory (isotopes of Te, Mo, Sr, Br, S, Sm, Ba, and Ti). Europium isotopes (masses 151 and 153) were separated in the calutron for the first time.

Approximately 30% of the calutron isotope separations time has been devoted to special types of separations involving attainment of the highest possible isotopic purity of both Li^6 and Li^7. Such purity requirements made mandatory the use of charge feeds already isotopically enriched. Certain development work was associated with these separations. Personnel responsible for development, design, fabrication, and evaluation of performance of calutron units were W. A. Bell, Jr., G. M. Banic, Jr., H. B. Greene, and W. K. Prater. Operations were performed under the direction of K. A. Spainhour, C. V. Ketron, R. L. Caldwell, W. W. Davis, and G. J. Fisher.

CALUTRON OPERATIONS

Tellurium, Series HC-XAX[1]

Collection of tellurium isotopes (Te^{120}, Te^{122}, Te^{123}, Te^{124}, Te^{125}, Te^{126}, Te^{128}, and Te^{130}) was started on April 6, 1955, and continued until June 6, 1955. Certain aspects of the collection – in particular, the retention of separated material in the collector pocket – were discussed briefly in the preceding semiannual report.[2] At that time, visual evidence indicated that a higher percentage of material would be retained by an aluminum receiving pocket than would be retained by a

[1] The suffix XAX (or XBX) indicates that separation was performed in equipment having a 48-in. (or 24-in.) collection radius, respectively.

[2] L. O. Love, *Stable Isotope Research and Production Semiann. Prog. Rep. May 20, 1955*, ORNL-1908, p 35 ff.

TABLE 11. RECOVERY OF TELLURIUM ISOTOPES FROM COPPER AND FROM ALUMINUM COLLECTORS

Isotope	Copper Collector			Aluminum Collector		
	Estimated Weight	Recovered Weight	Retention (%)	Estimated Weight	Recovered Weight	Retention (%)
Te^{120}	0.148	0.236	159	0.151	0.202	134
				0.083	0.045	54
Te^{122}	1.11	0.898	81	1.75	0.440	25
				0.749	0.430	57
Te^{123}	0.778	0.610	78	1.09	0.678	62
				0.512	0.318	62
Te^{124}	2.65	1.751	66	4.36	2.927	67
				1.72	1.042	60
Te^{125}	4.43	2.846	64	7.28	3.395	47
				3.28	1.934	59
Te^{126}	8.74	5.384	62	18.1	11.080	61
				6.67	3.770	56
Te^{128}	12.3	8.820	72	43.3	27.933	64
				13.8	9.44	69
Te^{130}	13.9	8.660	62	50.6	35.390	70
				15.7	12.316	78
Total	43.9			168.9		

pocket fabricated from copper, each pocket having comparable water-cooling. Subsequent recovery of the material in certain collector pockets shows wide variations in retention that cannot be explained.

The collected amount, in grams, of each isotope (estimated from meter readings) and the corresponding recovery of material from each type of collector are presented in Table 11.

A total of 2821 separator-hours was devoted to the tellurium series. Ion production was maintained for 2262 hr, during which period the total isotope beam current at the collectors averaged 19.8 ma. The peak ion current attained at the collectors was 32.6 ma.

Lithium, Series HD-XBX

This separation was started on May 23, 1955, to produce 25 g of Li^6 having, if possible, an isotopic purity of 99.999%. This material would be used in establishing primary assay standards and in fundamental research. A total of 3126 separator-hours was required in order to separate 35.5 g of Li^6. Of this amount, 23 g attained a purity in excess of 99.99%, 10 g of which assayed 99.999%.

The attainment of this Li^6 sample dictated the need for a "second-pass" type of operation, starting with LiCl charge feed prepared from material previously enriched in Li^6 content to 96.5%. Separation was performed in the 24-in.-radius calutrons by using the M-12 ion source equipped with a reflecting-anode-type arc chamber, $\frac{1}{8}$-in. 0-deg ion-exit slit, standard 0-deg accelerating electrodes, and 170-mil tantalum filament. Angular divergence, at the source, of the ion beams allowed to reach the receiver was limited to +11 to −7 deg. All lithium not actually entering the collector pockets was recovered, reprocessed, and again added to the charge feed.

The Li^6 and Li^7 collectors (Fig. 8) were designed and fabricated specifically for this collection. A water jacket, completely surrounding each

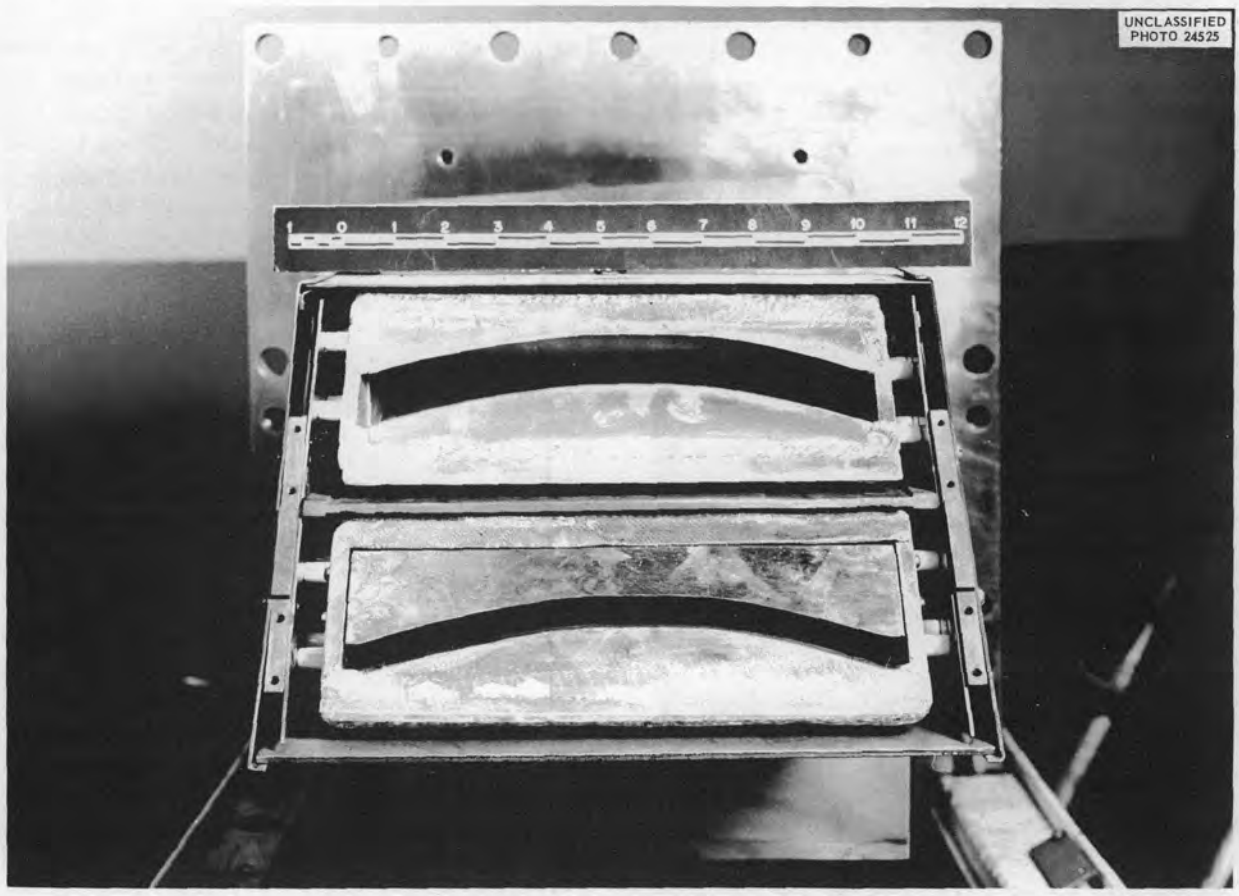

Fig. 8. Li^6 and Li^7 Collectors Positioned in Receiver.

pocket, provided extensive cooling of collected material. The size of the Li^6 pocket was chosen so that the point of impact of the Li^6 ions would be in the base or back portion of the collector, thus facilitating the removal of this material during acid leaching without contaminating it with lithium not deposited directly by the ion beam. In the completed receiver assembly, the Li^6 pocket was shielded mechanically from vapors by completely surrounding the pocket with a boxlike structure to ensure that the only entry into the pocket was through the $\frac{3}{16}$-in. beam-defining slot cut into the receiver faceplate. Ion entry into the Li^7 pocket was through a $\frac{1}{2}$-in. defining slot, made this size to reduce the amount of Li^7 which would strike the faceplate itself. To facilitate attainment of optimum ion-beam focus, a probe was mounted on the receiver faceplate in the $6\frac{1}{2}$ mass position to indicate ion scattering in that region. The assembled receiver, with the defining faceplate in position, is shown in Fig. 9.

Operational procedures were established with the main objectives of attaining the best possible beam focus and of performing the separation with the best possible process efficiency. The procedures adopted were as follows: pumpdown and outgassing time was extended before ion reception was attempted; nitrogen was used to support and stabilize the arc; ion output, at first limited to a maximum of 175 ma, was reduced later into the 100- to 125-ma range; tank surfaces were cooled with chilled water to condense neutral or unseparated lithium vapor; arc conditions were observed closely; and operating controls were never left unattended. At the end of each run the Li^6 collector pocket was removed from the receiver assembly immediately after its withdrawal from the vacuum chamber. Within minutes thereafter, the Li^6 deposited directly by the ion beam was leached from the pocket with dilute HCl, and an aliquot was taken for assay purposes. Material condensed in the remainder of the pocket was treated in like manner. Eventual combination of these small quantities (1 g or less) of material into a composite sample was made on the basis of assay results.

Fig. 9. High-Purity Li^6 Receiver with Beam-Defining Faceplate in Position.

A total of 62 runs was completed during the collection, 10 of which failed to produce ion beams. Ions were received for 2031 hr, during which time the total metered collector current averaged 77.3 ma.

During a survey of the collection, wide variations in purity were in evidence, but these variations had little or no correlation to operating parameters. Contamination of the separated Li^6 by normal lithium during the preparation and assaying of milligram quantities of the separated isotope is a possibility.

Lithium, Series HE-XAX

This separation was started on June 3, 1955, to produce 25 g of Li^7 having an isotopic purity of 99.99%. This material was to be used in conjunction with the Li^6 previously discussed in this report.

The ion source chosen for the collection was an Alpha-56 graphite-heater type of source. Collection was accomplished by using a 23.5-kv accelerating potential and a 2900-gauss magnetic field. The Li^7 ion beam then traveled an 8-in. radius and was focused at a point located some 4 in. in front of the source-receiver plane. Separation between the Li^6 and Li^7 beams at this point was 1.2 in. Magnetic focusing of these beams, by means of shims, was not attempted.

Charge feed used in the collection was a mixture of lithium plus LiCl in the ratio of 30 g of metallic lithium per 1000 g of LiCl. Isotopic concentration of Li^7 in each of the charge components was 97%.

Charge having this proportion of metal to salt was chosen because its use previously had been effective in preventing the formation of troublesome deposits which invariably formed on the arc side of the grid surfaces when LiCl alone was used as a charge.

The Li^7 receiving pocket was large, containing 450 in.^{2} of interior surface area well cooled by chilled water. Material collected in this pocket was divided into three parts: that where the ion beam impinged in the base of the pocket, that deposited on the top and ends of the pocket, and that deposited on the bottom of the pocket. Each division of material was leached from the pocket in the order listed above, immediately following removal of the pocket from the vacuum chamber. Assays were obtained before the final composite samples were made.

A total of 2061 separator-hours was devoted to the separation. Ion production was achieved for 1675 hr, with the production rate at the receiver averaging 229.8 ma. Peak production was 530 ma. The metered amount of Li^7 collected was 99.8 g. Thirty-five grams attained Li^7 purity in excess of 99.995%.

Molybdenum, Series HF-XAX and XBX

This separation was started on July 20, 1955, primarily to replenish the inventory of molybdenum isotopes (Mo^{92}, Mo^{94}, Mo^{95}, Mo^{96}, Mo^{97}, Mo^{98}, and Mo^{100}). During the collection three combinations of equipment were utilized to evaluate their usefulness in future molybdenum separation. The combinations were as follows: separation by use of Alpha equipment entirely, the source being a low-temperature two-arc modification of the Alpha-2 source; separation by use of a Beta M-12 source in the Alpha tanks; and separation in Beta equipment. In all three phases of the collection, $MoCl_5$ was used as charge material. Further evaluation of of MoO_3 as a charge was made in the graphite-heated type of Alpha-56 source unit. The above approach indicates clearly that previous experience with molybdenum separations has not been entirely satisfactory.

Attempts to use MoO_3 as a high-temperature charge in Alpha-ion sources met with failure. As soon as MoO_3 reached the arc region, decomposition to lower oxides occurred. Each of these oxides has a lower vapor pressure than does MoO_3, and condensation of these lower oxides on the ion-exit slit makes further operation impractical, if not impossible.

The $MoCl_5$ charge requires the use of a low-temperature (75 to 100°C) ion source. Temperature control in this range is often difficult to maintain, especially in the presence of varying drain conditions which contribute to the heating effects. Operating characteristics of $MoCl_5$ include: constant use of nitrogen support, severe drain to the negative electrode (especially with Alpha sources where electrode supports made of stainless steel melted), and formation of metallic molybdenum in pellet form within the arc chamber. Beam focus obtained with the Alpha source never was so good as that obtained with the Beta source equipped with comparable electrode geometry.

The isotopes were collected in water-cooled copper pockets, and the material was sampled in such a manner that the performance of each source may be checked against isotopic purity as soon as the product can be purified. The outputs obtained in this collection are presented in Table 12.

TABLE 12. COMPARISON OF MOLYBDENUM OPERATION WITH CHARGE-MATERIAL AND ION-SOURCE VARIATIONS

Ion Source	Tank	Charge Material	Separator Hours	Reception Hours	Average Ion Current (ma)	Peak Ion Current (ma)
Alpha-56, 2-arc	XAX	MoO_3	723	344	4.9	17.5
Alpha-2, 2-arc	XAX	$MoCl_5$	529	340	9.9	24.7
Beta M-12, 1-arc	XAX	$MoCl_5$	728	502	9.7	25.2
Beta M-12, 1-arc	XBX	$MoCl_5$	174	125	29.9	53.0

The following estimated amounts of molybdenum isotopes were collected:

Isotope	Amount (g)
Mo^{92}	4.99
Mo^{94}	4.08
Mo^{95}	7.29
Mo^{96}	8.21
Mo^{97}	5.72
Mo^{98}	13.3
Mo^{100}	5.67
Total	49.3

Strontium-Bromine, Series HG-XBX

The separation of strontium isotopes (Sr^{84}, Sr^{86}, Sr^{87}, and Sr^{88}) and of bromine isotopes (Br^{79} and Br^{81}) was performed simultaneously by ionizing $SrBr_2$ charge feed in a high-temperature (900 to 1000°C) Beta ion source. The collection was started on July 28 and continued through August 23, 1955.

The ion source was equipped with a $\frac{1}{8}$-in. 0-deg ion-exit slit, standard 0-deg accelerating electrodes, and a 170-mil tantalum filament. Strontium ions were received in water-cooled copper pockets. Bromine ions were received on magnesium chips contained in water-cooled copper pockets. All beam-defining slots were 0.125 in. in width. The collector pocket for Sr^{84} was placed in the reference position 48 in. from the ion-exit slit.

During operation, nitrogen was used to support the arc. Sparking flurries were frequent and unpredictable. Many runs were characterized by light "flick" sparking in the electrode region, which acted to trigger the flurries. In other runs, drains tended to creep, gradually building up and then becoming uncontrollable. Heater failures, both of the arc chamber and the charge oven, caused termination of 50% of the runs started. The majority of these failures occurred in the lead which connects the heater and the conductor because of motor action.

Magnesium chips in the bromine collector pockets were changed regularly; otherwise, evaporation of magnesium from the region subjected to ion bombardment left only bare copper in the path of the beam. The Sr^{88} collector eroded so badly that 14 such pockets were required for use during 702 hr of collection.

A total of 1210 separator-hours was devoted to this collection. Ions were received for 702 hr (58% of total time) at an average rate of 19.7 ma for strontium and 4.0 ma for bromine. Process efficiencies (metered amounts of isotopes collected/amount of element consumed, expressed as per cent) for strontium and bromine were 4.3 and 0.46%, respectively.

The following estimated amounts of each isotope were collected:

Isotope	Amount (g)
Sr^{84}	0.236
Sr^{86}	4.81
Sr^{87}	5.51
Sr^{88}	34.8
Total	45.3
Br^{79}	4.06
Br^{81}	4.01
Total	8.1

Sulfur, Series HH-XBX

Collection of sulfur isotopes (S^{32}, S^{33}, S^{34}, and S^{36}) was started on August 26, 1955, in order to replenish the inventory and to fulfill outstanding requests for S^{33}. Processing was performed with an M-12 ion source equipped with a reflecting-anode type of arc chamber having a $\frac{3}{8} \times \frac{1}{2}$ in. cross section, a $\frac{3}{16}$-in. 0-deg ion-exit slit, and standard 0-deg accelerating electrodes. The CS_2 charge was fed into the manifold region from a flask mounted outside the vacuum chamber. The rate of charge vapor flow was controlled by a leak valve of conventional type in which the packing around the stem was replaced with Teflon to give a more positive sealing effect against atmospheric leakage into the charge line. Nitrogen support was used in the arc at all times.

In previous collections of sulfur, only 10 to 20% of the S^{32} isotope had been retained in the collector pocket. The rejected material, in part, found its way into the S^{33}, S^{34}, and S^{36} collectors, as evidenced by retention in excess (as high as 150%) of that indicated from monitored currents. In this collection the sulfur ions impinged on copper filings contained in water-cooled copper collector pockets. Additional cooling passages were applied to the S^{32} pocket, and this pocket and the tank liner were cooled by chilled water. The S^{32} pocket was

shielded from the adjacent S^{33} collector by the placement of a formed stainless steel plate between these pockets. A similar plate was placed between the S^{33} and S^{34} pockets. Ion entry into the S^{32} pocket was through a 0.250-in. beam-defining slot. Similar slots 0.125 in. wide were provided for isotopes S^{33}, S^{34}, and S^{36}. The arrangement of receiver pockets and the shielding are shown in Fig. 10. The condition of the copper filings in each pocket and the amount of material rejected onto the receiver faceplate after 50 hr of ion reception also are indicated.

In operation a total ion beam of 60 ma was obtained in the first run. At a beam strength of approximately 30 ma, a greenish haze appeared around the receiver. This haze was progressively more dense as output was increased and indicated a greater loss of separated material. Consequently, output was limited to a maximum of 35 ma, and terminations were scheduled following every 50 hr of ion reception in order to remove and replenish the copper filings in the S^{32} pocket.

An S^{32} collector pocket was designed and fabricated in time to be used in the latter part of the collection. This pocket was much larger than the conventional collector, had more water-cooling passages, and had water-cooled fins or foot-scrapers on which it was hoped that the vaporized S^{32} would condense. During the time that this pocket was used, the output was increased to the 50- to 60-ma range. At this level a definite haze surrounded the receiver. The material has not yet been recovered from the pocket, but it is apparent that the pocket did not retain as much S^{32} as had been anticipated at the time of its design.

A total of 1486 separator-hours was devoted to this collection. Ion reception was maintained during 1241 hr at an average rate of 31.3 ma.

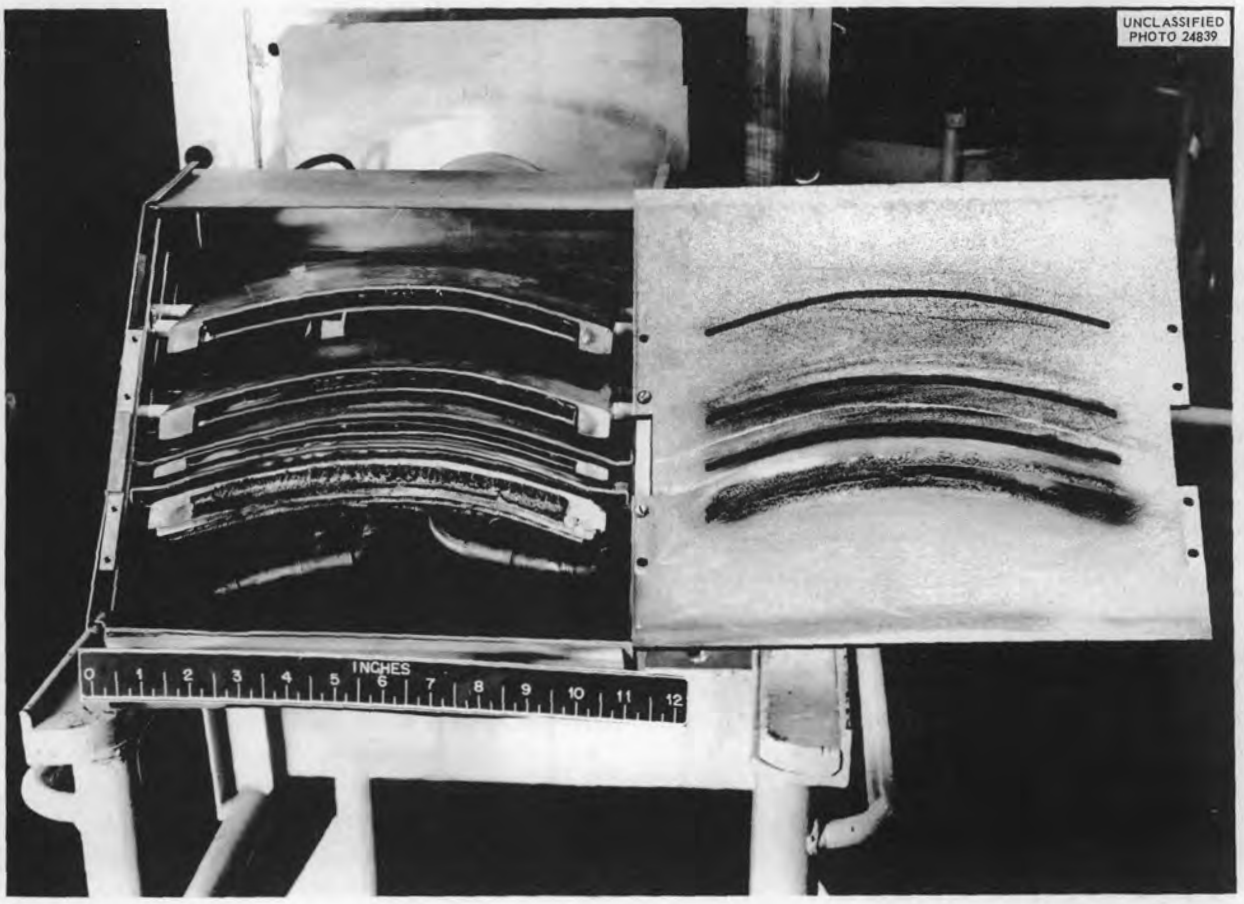

Fig. 10. Receiver Used in Collecting Sulfur Isotopes.

Process efficiency was 2.3%.

The following estimated amounts of sulfur isotopes were collected.

Isotope	Amount (g)
S^{32}	44.8
S^{33}	0.332
S^{34}	1.25
S^{36}	0.014
Total	46.4

Samarium, Series HI-XAX

This separation was started on August 31, 1955, to replenish the inventory of samarium isotopes (Sm^{144}, Sm^{147}, Sm^{148}, Sm^{149}, Sm^{150}, Sm^{152}, and Sm^{154}), with particular emphasis being placed on Sm^{150}.

Ions were produced in the M-14 ion source, which vaporized $SmCl_3$ charge at approximately 700°C. Collection of separated material was accomplished in purified-graphite pockets positioned behind suitable beam-defining slots.

Operation was characterized by heavy sparking, often in the form of uncontrollable flurries following slight changes in charge-oven temperature. The severity of sparking caused breakage of the negative electrode bushings and frequently caused the protective Lavite plates at the base of the positive bushings to melt. Adequate beam focus was extremely difficult to maintain. The focus was quite sensitive to arc changes or to changes in electrode position or potential, and the focal point frequently shifted to some value so much less than 180 deg that correct positioning of the receiver was impossible.

A total of 1031 separator-hours was devoted to this collection. Ion production, as metered at the receiver, was maintained for 617 hr at an average rate of 8.4 ma, the peak production being 19.4 ma. Filament life was longer than usual, averaging 45.8 hr per run.

The following estimated amounts were collected.

Isotope	Amount (g)
Sm^{144}	0.793
Sm^{147}	3.50
Sm^{148}	3.00
Sm^{149}	3.68
Sm^{150}	2.27
Sm^{152}	9.11
Sm^{154}	6.81
Total	29.2

Barium, Series HJ-XAX

The separation of barium isotopes (Ba^{130}, Ba^{132}, Ba^{134}, Ba^{135}, Ba^{136}, Ba^{137}, and Ba^{138}) was started on September 22, 1955, to replenish the inventory, particularly with isotopes Ba^{134}, Ba^{136}, and Ba^{137}.

Barium has always been considered a difficult element to process in the calutrons. Sparking is severe, and temperature control is extremely critical. During early stages of ion production, sparking is violent, gradually subsiding somewhat as optimum operating temperature is reached. Variation (either heating or cooling) from optimum charge temperature results in a prolonged series of sparking flurries.

In the present separation, evaluation of $BaCl_2$ as a charge in comparison with the normally used charge of metallic barium (99% Ba; 1% Sr) was contemplated. Actually, the comparison has been extended until, to date, the following charges have been used:

- $BaCl_2$,
- $BaCl_2$ plus metallic barium (96–98% Ba),
- metallic barium (96–98% Ba commercially packaged *in vacuo*),
- metallic barium (96–98% Ba commercially packaged *in vacuo* and opened and handled under oil),
- metallic barium (99% Ba commercially packaged in oil),
- metallic barium (commercial quality, distilled to 99.9% purity by members of this Division and handled under oil).

The main contaminant in all the above charges was strontium. As calutron charges, the first three [$BaCl_2$, $BaCl_2$ plus metallic barium, and metallic barium (96–98%)] were virtually inoperable. Sparking was so severe that source components failed, frequently before appreciable ion production could be attained. Satisfactory operation and reasonable separation rates were obtained only after the strontium content of the charge was reduced to 1%

(commercial purchase) or to less than 1% (distillation, reducing strontium concentration to less than 0.1%). Comparisons of operating parameters with each charge are tabulated in Table 13.

Separated isotopic material was collected in purified-graphite pockets positioned behind ⅛-in. defining slots. The pocket receiving Ba^{135} was machined so that it fitted tightly against the receiver faceplate, thus acting as a means of shielding the Ba^{134} from heavier masses rejected from their respective collector pockets.

This collection is nearing completion and the metered amounts of each isotope collected are as follows:

Isotope	Amount (g)
Ba^{130}	0.225
Ba^{132}	0.343
Ba^{134}	2.27
Ba^{135}	6.98
Ba^{136}	9.11
Ba^{137}	14.9
Ba^{138}	74.7
Total	108.5

Titanium, Series HK-XBX

The separation of titanium isotopes (Ti^{46}, Ti^{47}, Ti^{48}, Ti^{49}, and Ti^{50}) was started on September 26, 1955, with particular interest being in the collection of Ti^{50} to fulfill an outstanding request for that isotope.

The ion source used was identical to that used in the process described in the section titled "Sulfur, Series HH-XBX" in this report. Titanium tetrachloride charge was fed into the manifold region of the source from a flask mounted outside the vacuum chamber. Ion reception was in purified-graphite collector pockets.

The separation of titanium isotopes was comparatively simple in comparison with the other separations covered in this report. Operation was quite straightforward. Nitrogen was used to help support the arc, the focus was good, and equipment was used in run after run before major servicing was required.

Retention of separated material appeared to be less than desirable. The collected material was flaky and did not adhere firmly to the pocket walls. For this reason, receiving pockets were renewed periodically.

A total of 1009 separator-hours was devoted to this collection. Ion production was maintained for 818 hr at an average rate of 43.3 ma, the peak current at the receiver being 62.0 ma. Process efficiency was 12.0%.

TABLE 13. OPERATING PARAMETERS OF BARIUM CHARGES

Parameter	Charge					
	$BaCl_2$	$BaCl_2$ + Ba	Barium (96–98%)	Barium (96–98% in oil)	Barium (99% in oil)	Barium (99.9%, distilled)
Total runs	7	4	12	11	16	20
Number of runs producing ions	5	2	7	8	10	15
Separator hours	384	60	325	344	808	880
Ion-reception hours	188	10	94	162	468	456
Average current at receiver (ma)	5.4	1.7	6.1	9.5	22.3	19.0
Charge consumption of barium (g/hr)	3.5		3.4	1.4	1.3	1.3
Process efficiency (%)	0.7		0.9	3.3	8.1	7.0
Barium collected per separator hour (g)	0.013	0.001	0.009	0.022	0.063	0.048

The following estimated amounts of titanium isotopes were collected.

Isotope	Amount (g)
Ti^{46}	4.13
Ti^{47}	3.34
Ti^{48}	49.6
Ti^{49}	3.38
Ti^{50}	2.96
Total	63.4

Europium, Series HL-XBX

The first calutron separation of europium isotopes (Eu^{151} and Eu^{153}) was started on October 17, 1955, with the use of $EuCl_3$ charge produced by the Rare Earth Group of this Division. Since the oven temperature required to produce adequate charge vapor for ionization was higher than expected (1000°C), it is suspected that much of the charge became $EuCl_2$ before reaching the arc region. The material deposited within the arc chamber was a mixture of europium and $EuCl_2$.

This separation was performed in the Beta tanks, where adequate wash facilities for calutron components aid in the recovery of unseparated material. Ions were produced in the graphite-heated M-14 source, which was equipped with a $^3/_{16}$-in. 15-deg ion-exit slit, standard 0-deg accelerating electrodes, and a 170-mil tantalum filament. Ions were collected in purified-graphite pockets placed behind 0.125-in. defining slots. Material deposited in these pockets was in the form of hard flakes, which were easily lost unless the pockets were frequently replaced.

Operation was characterized by constant sparking and varying drain conditions. Output fluctuated almost constantly, frequently by as much as 100% within a period of minutes. Beam focus was never considered good, the beams being so broad that the separator between the two defining slots was maintained at a red heat. It became necessary to provide an expansion joint for this separator to prevent it from buckling and thus restricting the opening into the Eu^{153} collector.

A total of 1270 separator-hours was devoted to the collection. Ion reception was maintained for 664 hr at an average rate of 14.6 ma, the maximum rate being 56.0 ma. Process efficiency was 5.65%.

The following estimated amounts of europium isotopes were collected.

Isotope	Amount (g)
Eu^{151}	27.2
Eu^{153}	28.0
Total	55.2

DEVELOPMENT AND PROCESS IMPROVEMENT

W. A. Bell, Jr. H. B. Greene
G. M. Banic, Jr. W. K. Prater

There is associated with calutron seperations a development program and a process improvement program. In many aspects, the two are so closely related that a clean-cut separation between them is highly impractical in that equipment generally is not taken out of a production state while new ideas are being evaluated.

In the past six months the projects with which members of the Development Group have been associated are listed as follows.

XAX Motor-Generator Installation and Proposed Magnet Expansion

Installation of the 500-kw, 250-v, 2000-amp motor-generator is progressing according to schedule and should be completed by December 19, 1955. This machine, when used to excite the XAX magnet, will make it possible for a magnetic field intensity to be obtained in the vicinity of 5000 oersteds. When the magnetization curves are completed, they will be plotted to determine the field distribution and maximum intensity.

The engineering required for the expansion of present XAX isotope separation facilities from two to four calutrons is approximately 75% complete. The required engineering includes:

1. removing the existing magnet and coils, two Alpha-2 type of bins, manifolds, and 20-in. diffusion pump assemblies, and one Alpha-I type of high-voltage power supply;
2. relocating two circulating oil pumps, two Kinney vacuum pumps, two vacuum scrubbers, and one hot water heater and pump, with their associated piping, valves, and controls; all hot, cold, and chilled water lines; two magnet

excitation buses; and several miscellaneous utility services;

3. increasing the architectural structures that are required to support the load imposed by the modified yoke and the additional coils and tanks;
4. reassembling the magnet yoke, rotated 90 deg from its present position and containing 18% more iron;
5. installing the three existing and two additional coils and core materials; four Alpha-2 type of bins, manifolds, and 30-in. diffusion pump assemblies; three additional Kinney pumps; all associated piping, valves, and controls; and two additional Alpha-2 type of power supplies with control equipment and connecting cables.

Low-Temperature Multiple-Arc Alpha-2 Source

For some time, there has been a need for a large multiple-arc calutron source which could be used to process charge materials that require oven temperatures in the 50 to 150°C range. An Alpha-2 source was modified to accommodate these charge temperatures and was used in processing the isotopes of molybdenum from $MoCl_5$ charge.

The major modifications required to lower the Alpha-2 operating range to this extent included the elimination of two (Nos. 2 and 4) of the four arc chambers initially provided, the insertion of a water-cooled pad and several layers of stainless steel heat shielding between the charge heater castings and their respective manifolds, the replacement of the heating element in the top of each charge casting with a water-cooling coil soldered in place, and a reduction in the total number of heating elements used. Extension of the temperature range may be accomplished by the use of more heaters and/or reduction in the amount of water-cooling applied.

DEVELOPMENT

W. A. Bell, Jr. H. B. Greene
G. M. Banic, Jr. W. K. Prater

Investigation of the Entrance of Neutral Particles into Receiving Pockets

In collections involving the attainment of high-purity samples, contamination by neutral vapors creates a problem. One means of reducing neutral contamination consists in cooling or chilling the walls of the tank liner to condense a large portion of these unwanted vapors. In lithium separation a single attempt was made to evaluate the amounts of neutral material entering the collector pockets. Three collector pockets which were as nearly identical in size and cooling as was possible were installed. One was a control pocket located in the $5\frac{1}{2}$ mass position; the other two pockets were for the reception of lithium masses 6 and 7. Pockets in the $5\frac{1}{2}$ and 6 mass positions were placed one directly above the other and were positioned behind identical beam-defining slots 0.187 in. in width. The pocket for reception of mass 7 was in its normal position relative to the mass-6 collector and was preceded by a 0.500-in. beam-defining slot. Lithium separation from Li^6Cl charge (96.5% Li^6) was performed while $FeCl_2$ was vaporized from an auxiliary oven located near the lithium ion source. This $FeCl_2$ vapor was not ionized before being released into the tank region. The data in Table 14 are the results obtained in this single experiment. It is interesting to note that approximately ten times as much iron was found in the Li^6 collector as was found in the control pocket, which did not receive an appreciable ion beam. The presence of more lithium than iron in the control pocket probably indicates ion scattering, since approximately twice as much iron as lithium was liberated into the vacuum chamber.

TABLE 14. RECOVERY OF LITHIUM RELATIVE TO NEUTRAL-IRON CONTAMINATION

Collector	Average Ion Current (ma)	Lithium (mg)	Iron (mg)	Assay (% Li^6)
Li^7	2.2		1.5	
Li^6	67.1	1672.0	1.8	99.929
Control	0.0	1.05	0.14	

High-Purity Lithium Collector (Beta)

The purity restrictions (99.999% Li^6) placed on lithium separation made retention of all collected material mandatory; if retention of the material had not been accomplished, dilution of the sample by rejected Li^7 might have occurred. The problem

of increasing the retention of lithium was approached directly by applying to each collector all the water-cooling possible, using a pocket as large as receiver space limitations would permit. The assembled receiver (with faceplate removed) is shown in Fig. 8. Each pocket is surrounded on top, bottom, and back with approximately 10 ft of water-cooling passages which are $\frac{3}{16} \times \frac{1}{2}$ in. in cross section and are separated by spaces of $\frac{1}{8}$ in. Pocket-wall thickness from the interior surface to the water-cooling passages is 0.100 in.

Extensive Cooling of Conventional Beta-Receiving Pocket

The lithium collectors mentioned above were so effective in retaining lithium that the same cooling technique was applied to some conventional Beta copper collectors. Water-cooling passages were milled into $\frac{1}{4}$-in. copper stock, which was then welded onto the already fabricated pocket. The addition of two such cooling pads to each collector supplied that pocket with approximately 8 ft of cooling passages and added only $\frac{1}{2}$ in. to the thickness of the pocket.

Alpha-2-Source Negative-Electrode Adjustment

In Alpha-source design, whether or not the negative electrode can be positioned properly is of major uncertainty, expansion of components and other factors being as they are. A rather cumbersome – but workable – electrode adjustment mechanism was attached to the ground frame of the Alpha source between the high-voltage insulators. Relocation of the electrode support insulators on a sliding mechanism activated by an off-center cam provided sufficient movement to verify the need for movable negative electrodes on Alpha equipment.

Alpha 20-in.-Radius Receiver Adjustment

Collections of lithium and calcium have been made at a 20-in. collection radius in Alpha tanks. In the lithium collections, receivers mounted rigidly in the liner proved to be quite adequate. Locating the receiver in its optimum position for the more closely spaced calcium isotopes imposed a more difficult problem. The need for adjustment of the receiver perpendicular to the source-receiver plane was in evidence. A supporting frame which will permit receiver adjustments to be made has been designed, fabricated, and used.

"Nonleak" Beta Charge Bottle

A review of calutron operations reveals that process efficiency varies from 2 to 20%. Part of this variation is attributable to leakage of charge material where imperfections exist in the seal between the charge bottle and the manifold orifice. Both flat and spherical surfaces had been used at this location, often quite successfully; however, occasional leaks would occur. In collections involving high-purity samples or when charge material is precious, leaks cannot be tolerated. Such was the case in high-purity lithium separation. For this separation a conical seal was designed and was used so successfully that its use is being extended to include other separations. The modified components are shown in Fig. 11.

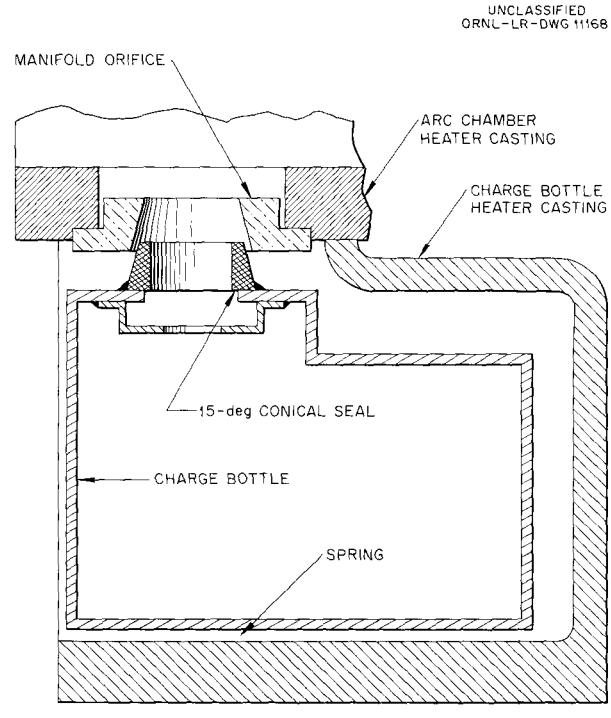

Fig. 11. Beta Charge Bottle with Conical Shield.

Adjustable 90-deg Baffle for Beta Tanks

Limiting the angular divergence of ion beams reaching the receiver by means of movable baffles in the 90-deg position is made possible while the tank is in operation by using push-rods that extend through the liner faceplate. Such an arrangement,

in addition to limiting total angular beam divergence, allows the choice of any series of diverging angles lying between + 13 and – 11 deg.

Expansion Slot in Beta Receiver Faceplate

It has been generally noted that the edges of the ion-beam-defining slot which precedes a collector pocket become heated whenever beam currents are in excess of 30 to 40 ma. If the spacing between adjacent ion beams is less than, say, ½ in., this heating causes expansion of the thin graphite strip that forms the separator between defining slots. This strip, being an integral part of the graphite faceplate itself, then buckles in the middle and reduces the opening into one of the collector pockets. The introduction of an expansion joint at one end of the separating strip has prevented this buckling or distortion in both barium and europium separations.

Improvements to Carbon Tube Furnace

The purification of graphite collector pockets in the carbon tube furnace seems to be a never-ending job, and much time is required to keep the furnace in repair. Components which failed have been replaced with improved parts whenever such changes could be incorporated without major overhaul of the entire furnace. Electrode burnout and breakage of the 6-in.-dia tube have been common failures. Improvements in the expansion linkages and in the contacts between electrodes and water-cooled metal conductors have been incorporated recently into the furnace design.

Increase in Electron Dumping Areas in Barium Separation

Barium separations are characterized by sparking flurries and high drains – particularly the drain from the negative electrode to the positive supply. Such drain conditions drive the negative electrode positive with respect to ground and cause the supporting insulator to crack. One ion source was modified to increase the area and location of electron dumping areas maintained at high positive potential and to minimize the regions where drain from the negative to positive regions could occur. Such a drain system did not produce detectable improvement in operating conditions; however, points of localized heating were reduced and insulator failures were retarded.

Improvement in Methods of Fabrication of Graphite Receiver Pockets

The fabrication of graphite receiver pockets having $\frac{1}{16}$-in. wall thickness and $\frac{3}{16} \times 4$ in. interiors has been a slow and tedious process. Formerly, these pockets were fabricated by a method in which a few thousandths of an inch of graphite was removed with each passage of the cutting tool. Recently, the Stable Isotope Group undertook direction of graphite pocket fabrication. The bulk of the graphite now is "hogged out" by drilling before milling operations are started. Pocket fabrication cost has been greatly reduced and fabrication time has been reduced by almost a factor of 8.

Status of Inert Gas Collection (Argon)

Experimentation has continued in connection with the collection of inert gases in the calutron. Efforts have been directed toward the adaptation of the method reported[3] into a practical, usable system. This method consists in continually or periodically condensing magnesium vapor upon the target on which the ions of the inert gas (argon) are impinging.

A major problem in this method is that of obtaining controlled vaporization of magnesium at a rate of approximately 0.1 g/hr. The use of various sizes and types of furnaces, all containing magnesium metal (again in various sizes and having been given various surface treatments), has failed to produce a satisfactory vaporization rate. It was always necessary to start the vaporization at some temperature higher than optimum and then to reduce the temperature once vaporization was established. Even then, optimum furnace temperature varied from 390 to approximately 460°C.

A more satisfactory arrangement for magnesium vaporization consists in feeding magnesium ribbon into the center of a heated tantalum coil. With this method, difficulties arise from the motor action created by the magnetic field in which the heated coil must be placed, and from the condensation of magnesium upon certain exposed parts of the feeding mechanism. The first problem was solved by the selection of proper coil size, use of direct current for heating the coil, and proper orientation of the coil with respect to the magnetic

[3] P. S. Baker, F. R. Duncan, and H. B. Greene, *Stable Isotope Research and Production Semiann. Prog. Rep. May 20, 1955*, ORNL-1908, p 45.

poles of the calutron. The condensation of magnesium on the exposed end of the ribbon guide has always been the limiting factor in operation. The mechanism itself, in which a bellows is pulsed periodically to activate a ratchet, thereby moving the magnesium at a controlled rate, functions quite nicely. Failure to feed occurs only when condensed magnesium "freezes" the ribbon being pushed into the heated coil.

Several collection runs of short duration have been completed in the Beta channels. Output of A^{40} reached 44 ma, with the A^{36} output being 0.05 ma. No attempt was made to collect A^{38}. Monitored currents indicate that 0.0025 g of A^{36} impinged on the collector. The trapped A^{36} has been recovered in two portions, that from where the more intense portion of the beam had impinged and that which was trapped in the remainder of the target. Retention of argon was in excess of 50%, and the purity of the two samples was 36 and 11% A^{36}, respectively.

Design of Receiver Pocket for High-Purity Li^6 Collection Using Alpha Source

In separating lithium the focal points of the ion beams are shifted some 4 in. forward from the 180-deg position. The "second-pass" separation using enriched Li^6 charge material requires a collector for Li^6 which can dissipate 10 to 15 kw of energy. Moreover, the location of this collector relative to the location of the Li^7 collector dictates that the Li^6 collector must have thin walls and a rather peculiar shape. This collector, its required insulators, and support structures are now in the Machine Shop for fabrication.

STABLE ISOTOPE CHEMISTRY

Boyd Weaver, Department Head

INTRODUCTION

Charge materials were prepared for the separation of isotopes of eight elements. Chemical refinement of isotopes of seven elements was completed. Activities in the separation of rare earths included the preparation of Y_2O_3 of extremely high purity, the production of more than 500 g of Eu_2O_3, the improvement of Dy_2O_3 for charge material, and the preliminary processing of more than 100 kg of gadolinite ore.

PREPARATION OF CHARGE MATERIALS

C. W. Sheridan W. C. Davis

Materials were prepared for the separation of the isotopes of eight elements: S, Ti, Sr, Br, Sm, Eu, Mo, and Ba. Charge materials for sulfur and titanium were commercial CS_2 and $TiCl_4$, and the isotopes of strontium and bromine were separated simultaneously by using commercial $SrBr_2$.

The chlorides of samarium and europium were prepared by heating the oxides with NH_4Cl and removing the excess NH_4Cl by heating *in vacuo*. Both $SmCl_3$ and $EuCl_3$ prepared in this way were completely soluble in water, indicating the absence of oxychlorides. After the calutron operations, the samarium which had not been collected as separated isotopes was recovered from various parts of the calutrons, chemically purified, and returned to the rare-earth storage.

As a result of the rare-earth separation activities of the department, there was available for europium charge material approximately 625 g of Eu_2O_3, equivalent to 915 g of $EuCl_3$. By rapid recovery and chemical processing of material deposited on calutron parts, this quantity was extended effectively to 1750 g of $EuCl_3$. Volatilization of $EuCl_3$ in a charge bottle is accompanied by thermal reduction and dissociation. The vapor introduced into the ionizing chamber is largely $EuCl_2$. The residue in one charge bottle had an average composition of $EuCl_{2.2}$, indicating that reduction takes place in the solid state. The chlorine set free is very corrosive to stainless steel charge bottles.

The recycle of europium charge material was accomplished by washing calutron parts, filtering the solutions, precipitating the samarium as hydroxide and oxalate, igniting the oxalate to oxide, and converting it to chloride. The impurities which remained did not interfere with calutron operations. Since the completion of the isotope separation, a final recovery is in process in which many kilograms of graphite parts are being burned and the very valuable europium is subjected to a rigorous purification before storage as oxide.

Ten kilograms of $MoCl_5$ was prepared by direct synthesis of the elements at 525°C. Platinized asbestos was found to be an effective catalyst for the reaction. The product distilled from the reactants and was collected in a cool part of the reaction tube.

Chemical operations in the preparation of barium charge material consisted in purifying the metal by fractional distillation of the slightly more volatile strontium, which is present in all commercial barium. It was usually found necessary to distill away at least 75% of the barium in order to sufficiently purify the remainder.

Operational experience with all these charge materials is given in more detail in the section titled "Calutron Operations."

CHEMICAL REFINEMENT OF ISOTOPES

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F. B. Thomas F. M. Scheitlin

Progress was made in the chemical refinement of ten elements: N, Ni, Te, Rb, Ru, Ca, Sr, Li, Br, and Mo. The following discussion concerns only those elements which have been processed through complete series.

Nitrogen

For the collection of enriched N^{15} the collector pockets were filled with metals chosen because of the possibility that they would collect the active nitrogen in a beam but would not collect the neutral nitrogen. The metals used as collectors were magnesium, aluminum, sodium, and uranium. Procedures for recovery of the minute amounts of nitrogen collected were dependent upon the metals used.

Magnesium turnings used as a collector were placed in a flask and allowed to react with a solution of NaOH. Any nitrogen present as nitride was reduced to NH_3, carried by the large amount

of H_2 produced and a stream of argon, and collected in dilute HCl. The NH_4Cl was dried carefully on a steam bath. In two separate experiments, each involving about 100 g of magnesium, the weights of the residues were 3 mg in one case and 1 mg in the other. These residues were not all NH_4Cl.

About 100 g of aluminum used as a collector was treated in the same manner as the magnesium, giving 6.6 mg of NH_4Cl.

Sodium used as a collector was treated slowly with water and, finally, with steam, without yielding a weighable quantity of NH_4Cl.

Uranium used as collectors was fused with NaOH, and the gases evolved were carried by argon into dilute HCl. Two collections of NH_4Cl weighed 2.8 and 30 mg. A collection of N^{14} estimated as 2 g yielded only 96 mg of N_2.

Nickel

The procedure used for refining one series of nickel isotopes depended upon the amount to be processed.

For quantities less than 1 g the following procedure is used:

1. Ignite carbon collectors at 650°C in O_2.
2. Dissolve ash with HCl and H_2O_2, dilute, and filter.
3. Make alkaline with NH_4OH, digest, and filter.
4. Acidify filtrate with CH_3COOH.
5. Precipitate Ni with ethanol solution of dimethylglyoxime and filter.
6. Dissolve precipitate with 6 *N* HNO_3.
7. Dilute and electrolyze with Pt electrodes at 1 amp.
8. Add H_2SO_4 and evaporate to dryness.
9. Dissolve in H_2O, filter, and evaporate to dryness.
10. Ignite to NiO in silica ware at 900°C.

For quantities greater than 1 g the following procedure is used:

1. Ignite carbon collectors at 650°C in O_2.
2. Dissolve ash with HCl and H_2O_2, dilute, and filter.
3. Make alkaline with NH_4OH, digest, and filter.
4. Acidify filtrate with H_2SO_4.
5. Saturate with H_2S, digest, and filter.
6. Evaporate filtrate to dryness, dissolve in H_2O, and filter.
7. Evaporate filtrate to dryness and ignite to NiO in silica ware at 900°C.
8. Digest with boiling H_2O, filter, dry, and ignite in silica ware at 900°C.

Production data for the series are in Table 15.

Tellurium

Collection of tellurium isotopes on copper has been found to be very inefficient. Therefore during the latest separation series aluminum collectors were substituted. The whole series resulted in a total of 31 lots in four groups: (a) collections on copper, (b) and (c) collections on aluminum, and (d) material falling from (a) and (b) collectors to the tops of the next lower collectors. The following procedures were used in the refinement of these lots:

1. Dissolve Te from collectors with HNO_3. (Aluminum requires aqua regia.)
2. Evaporate to dryness on a steam bath.
3. Add HCl and evaporate to strong fumes.
4. Repeat HCl treatment twice.
5. Dilute to 3 *N* HCl, add hydrazine hydrochloride, and heat.
6. Introduce SO_2 until Te precipitates. Filter and add more SO_2 to filtrate. Reduction from

TABLE 15. PRODUCTION DATA ON NICKEL ISOTOPES

Isotope	Lot No.	Weight of NiO (g)	Impurities Found (%)							
			Co	Cu	Fe	Mg	Na	Si	Mn	V
58	GT 844(a)	174.480		0.2	<0.02	0.1				
60	GT 845(a)	61.347	<0.05	<0.05		<0.02	0.01			0.02
61	GT 846(a)	1.324		0.05	0.02	0.01		0.01		
62	GT 847(a)	7.377				<0.02				0.02
64	GT 845(a)	1.211		0.05	<0.02				<0.01	

copper solution requires much more reducing agent because much of the agent is used in reducing Cu(II) to Cu(I), which is continually reoxidized by air.

7. Add about 1 g of $Fe_2(SO_4)_3$, make alkaline with NaOH, and filter $Fe(OH)_3$ with residual Te. In copper solutions, $Al(OH)_3$ is precipitated by NH_4OH as a scavenging agent.

8. Dissolve precipitate with HCl and reduce again with $N_2H_4 \cdot 2HCl$ and SO_2.

9. Combine Te fractions, dissolve in 1:1 HNO_3, convert to chloride solution, and repeat reduction.

10. Wash Te in centrifuge tube first with warm H_2O, then with acetone.

11. Dry at 110°C.

The isotopes of group (d) – see the preceding paragraph – were accompanied through all stages by some unknown organic matter. It was finally removed by filtering the HCl solutions of tellurium through decolorizing charcoal. Production data are in Table 16.

Rubidium

Two rubidium isotopes (85 and 87) were refined by the following procedure:

1. Dissolve Rb from Cu collectors with 1:1 HCl.
2. Evaporate to dryness, dissolve in very dilute HCl, and filter.
3. Electrolyze filtrate, using carbon sheet electrodes, and filter.
4. Make alkaline with NH_4OH, introduce H_2S, and filter.
5. Acidify slightly with HCl, boil, precipitate with $BaCl_2$, and filter.
6. Evaporate filtrate to dryness and transfer to silica dish.
7. Fuse RbCl.
8. Dissolve in H_2O, evaporate to dryness, redissolve, and filter.
9. Remove Ca by NH_4OH and $(NH_4)_2CO_3$ precipitation.
10. Acidify with HCl and evaporate to dry RbCl.
11. Remove Na and K by elution from a 3-ft column of Amberlite IR-100 with 0.1 *N* HCl.

In the present instance the alkali separation was performed by elution from Dowex-50 with 0.1, 0.3, and 0.5 *N* HCl. Only the last removed the Rb from the resin, and it did not give good separation. Consequently, the isotopes are divided into fractions. Production data are in Table 17.

SEPARATION OF RARE EARTHS

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R. L. Bailey W. C. Davis

Most of the effort in the separation of rare earths was applied to the production of europium for calutron separation of Eu^{151} and Eu^{153}. About 500 g of Eu_2O_3 was added to the previous production of 200 g. Operation of the continuous-countercurrent tributyl phosphate–nitric acid extraction process for the separation of other rare earths had previously resulted in various fractions containing from 0.5 to 5% Eu_2O_3, mixed with Sm_2O_3 and Gd_2O_3. These fractions were subjected to further extractions in such a way as to concentrate most of the europium into smaller fractions containing from 5 to 50% Eu_2O_3.

Treatment of mixed rare-earth acetates with sodium amalgam gave a europium-samarium amalgam and a gadolinium byproduct containing 0.2% europium. Washing the mixed amalgam with dilute H_2SO_4 precipitated $EuSO_4$ and left most of the samarium in solution. The $EuSO_4$ was oxidized with HNO_3 and converted, via oxalate precipitation, to the oxide. The Eu_2O_3 had purities of from 70 to 95%, depending largely on the original samarium content.

Further purification of the europium was obtained by a second amalgamation followed by precipitation as $EuCl_2$ from 12 *N* HCl. This stage is not quantitative, but the residual mixture is available for reprocessing. In the last large-batch operation of this process, 125 g of 95% Eu_2O_3 was obtained from the sulfate precipitation. The final treatment gave 106 g of less than 99.95% Eu_2O_3. Approximately 1500 lb of 0.25% sodium amalgam was consumed in the production of 700 g of Eu_2O_3. This is by far the largest known quantity of europium in existence.

A laboratory-scale extraction system is being used to separate rare earths from other elements in gadolinite, an ore containing about 40% Y_2O_3 and rare earths, along with beryllium and iron silicates. The crushed ore is opened by leaching with HNO_3, which dissolves all but the silica. The feed solution to the extraction section of the column system is 15 *N* in HNO_3 and contains about 100 g per liter of earth oxides. The stripping section is fed at an equal rate, 1.2 liters/hr, with 6 *N* HNO_3. Combination of the two flows gives

10.5 *N* HNO_3 in the extraction section. A countercurrent flow of 2.5 liters per hour of tributyl phosphate extracts out the yttrium and heavy lanthanons, leaving the 15% content of cerium earths in the aqueous effluent along with the iron and beryllium. Approximately 50 kg of oxide has been separated thus far.

An 11-stage mixer–settler has been constructed and is being operated to obtain fundamental information on the stage-to-stage behavior of yttrium earths in the extraction process.

Attempts were made to improve the quality of approximately 1 kg of 90 to 95% Dy_2O_3 previously produced by liquid-liquid extraction. Batch elutions were made from both hot and cold cation-exchange columns with 5% citric acid. Difficulties were encountered from crystallization when as much as 150 g of Dy_2O_3 was loaded on a 3-in. column.

TABLE 16. PRODUCTION DATA ON TELLURIUM ISOTOPES

Isotope	Lot No.	Weight of Te (g)	Impurities Found (%)											
			Ag	Al	Ba	Cr	Cu	Mg	Mo	Mn	Na	Ni	Si	Sn
120	HC 875(a)	0.236			<0.02								<0.05	
120	HC 875(b)	0.202					0.01			<0.01			0.02	0.1
120	HC 875(c)	0.051					<0.05						0.05	
122	HC 876(a)	0.898					No impurities detected							
122	HC 876(b)	0.440	Trace				<0.02	<0.02					0.05	
122	HC 876(c)	0.430											<0.05	
122	HC 876(d)	0.240					No impurities detected							
123	HC 877(a)	0.610					<0.05	<0.02	<0.02					
123	HC 877(b)	0.678	Trace				0.05						0.05	
123	HC 877(c)	0.318	Trace					<0.02					0.2	
123	HC 877(d)	0.235					<0.05							
124	HC 878(a)	1.751											<0.02	
124	HC 878(b)	2.927	Faint trace				<0.05	<0.02					<0.05	
124	HC 878(c)	1.042											<0.02	
124	HC 878(d)	0.652			0.05			0.02			<0.01		0.05	
125	HC 879(a)	2.846					<0.05	<0.02						
125	HC 879(b)	3.395	Trace				0.05						<0.05	
125	HC 879(c)	1.934	Trace				<0.01						0.1	
125	HC 879(d)	1.002				0.08						0.08	<0.02	
126	HC 880(a)	5.384					No impurities detected							
126	HC 880(b)	11.080					<0.05						<0.05	
126	HC 880(c)	3.770					<0.05						<0.05	
126	HC 880(d)	2.881						0.01					<0.02	
128	HC 881(a)	8.820	Trace				<0.05				<0.01		<0.05	
128	HC 881(b)	27.933					<0.05	<0.02			0.05		<0.05	
128	HC 881(c)	9.440					<0.05						<0.05	
128	HC 881(d)	5.182											<0.02	
130	HC 882(a)	8.660					0.02							
130	HC 882(b)	35.390									0.01		<0.05	
130	HC 882(c)	12.316		<0.05			<0.05				<0.01		0.1	
130	HC 882(d)	5.501											<0.02	

TABLE 17. PRODUCTION DATA ON RUBIDIUM ISOTOPES

Isotope	Lot No.	Weight of RbCl (g)	Impurities Found (%)					
			Ca	Cu	K	Li	Mg	Na
85	GX 862(a)	47.413	0.2		0.3	<0.01	0.1	0.2
87	GX 863(a)	2.121	0.2		2.0	0.002	0.05	0.1
87	GX 863(a-2)	2.891	0.1	0.1	0.3	0.2	<0.02	0.2
87	GX 863(a-3)	4.898	0.2	0.1	0.1	0.1	0.02	0.3

Heavy loading of heated columns resulted in breakage of the column at the base, apparently because of expansion of the resin in changing from the hydrogen form to the ammonium form. A considerable part of the terbium and holmium impurity was removed from several fractions. There is on hand more than 600 g of Dy_2O_3 better than 95% pure and small quantities at the 99 and 99.9% levels. This will permit isotope separation on a recycle basis. Extraction had removed essentially all the gadolinium, the most undesirable impurity in the separation of dysprosium isotopes.

PURIFICATION OF YTTRIUM

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R. L. Bailey W. C. Davis
J. A. Norris[1] J. R. Walton[2]
L. E. Stanford[3]

The Chemistry Department of this Division took part in a project to obtain sufficiently pure yttrium so that its cross section for absorption of slow neutrons could be determined accurately. Since dysprosium, usually the most abundant impurity in highly purified yttrium, has a cross section of about 1100 barns, and samarium and gadolinium have cross sections of 6,500 and 44,000 barns, respectively, extremely small quantities of these elements can have significant effects on the cross section of yttrium, already known to be less than 1.5 barns.

The starting material consisted of two batches of yttria already purified to a high degree at Ames Laboratory by the process developed there, which involves elution from a system of cation-exchange columns by ethylenediaminetetraacetic acid at pH 8. These batches are designated hereinafter as A-1 and G.

The progress made in the chemistry laboratory was followed by spectrochemistry, mass spectrometry, and cross-section determinations in the oscillator of the ORNL Graphite Reactor. Spectrochemical measurements did not detect samarium nor gadolinium in batch A-1, but, after chemical fractionation, spectrochemical analysis of the fractions in which these elements were thus concentrated indicated that the original samarium content was less than 5 ppm and that the original gadolinium content was less than 0.5 ppm. Analysis of batch G showed 80 ppm samarium and noticeable quantities of dysprosium, gadolinium, and even lanthanum. Mass spectrometry detected traces of lanthanum, cerium, and samarium and a faint indication of gadolinium in A-1, while G was found to contain larger amounts of lanthanum, cerium, samarium, gadolinium, and dysprosium and a trace of neodymium. The best cross-section values for A-1 and G were 1.55 and 2.36 barns, respectively.

Initial plans for purification were based on indications that the most significant impurity would be dysprosium. The process of slowly increasing the pH of a solution of yttrium and lanthanon nitrates by introducing air-borne ammonia precipitates the heavier lanthanons preferentially. Therefore, nitrate solutions of three different concentrations were each divided into several successive fractions by this method. In each case the final fraction was found to have a greatly reduced dysprosium content, but the samarium content was

[1] J. A. Norris, Spectrochemical Research Laboratory.

[2] J. R. Walton, Mass Spectrometry Laboratory.

[3] L. E. Stanford, on loan from G.E.

increased sufficiently to overcome any decrease in the cross section.

A 50-g quantity of A-1 was loaded on a large cation-exchange column and eluted with 5% citric acid at pH 3 to give 15 fractions. The middle fraction gave a cross section of 1.47 barns. Samarium and gadolinium were detected in the final two fractions.

Separate treatment of two samples of G in acetic acid with sodium amalgam decreased the samarium content from 80 to less than 30 ppm and gave cross sections of 1.82 and 1.50 barns. Thus samarium appeared to be the most significant neutron absorber among the impurities in G. This process can remove only samarium and the much rarer elements europium and ytterbium.

Fractional precipitation of rare-earth oxalates from homogeneous solution by hydrolysis of methyl oxalate precipitates samarium preferentially, with the other elements on either side following progressively. Yttrium tends to remain behind all the rare earths, with the possible exception of lanthanum and cerium. By this means 50 g of G and 100 g of A-1 were divided into several fractions. Analyses are given in Tables 18 and 19. There is an apparent anomaly in the cross sections of A-1, where the values decrease toward a limiting value, then abruptly increase before again decreasing asymptotically. Since the gold used for calibration was observed to behave similarly, it is possible that all fractions from A-1 beyond the fourth were of essentially the same quality. Mass spectrometric examination of the final fraction from A-1 showed traces of lanthanum and cerium but no other lanthanons.

On the basis of the evidence presented above, it is concluded by members of this division that the cross section of yttrium is approximately 1.40 barns, in close agreement with a previous value[4] of 1.38.

[4]D. J. Hughes and J. A. Harvey, *Neutron Cross Sections*, BNL-325 (July 1, 1955).

TABLE 19. METHYL OXALATE FRACTIONATION OF MATERIAL A-1

Fraction	Oxide Weight (g)	Cross Section (barns)
1	8.91	1.75
2	10.64	1.60
3	10.81	1.56
4	10.93	1.45
5	11.87	1.41
6	11.57	1.56
7	8.99	1.49
8	9.33	1.45
9	10.80	1.39
10	4.79	1.41
	98.64	

TABLE 18. METHYL OXALATE FRACTIONATION OF MATERIAL G

Fraction	Oxide Weight (g)	Impurities Determined (ppm)*			Cross Section (barns)
		Dy	Sm	La	
1	7.03	430	275	90	3.08
2	5.25	285	<100	90	
3	5.53	330		115	
4	5.42	215	50	95	
5	6.05	190	60	95	
6	5.14	150		130	
7	9.84	90		110	
8	5.77	30		175	1.45
	50.03				

*Parts per million by weight of oxide to oxide, error is estimated at ±20% of amount listed; blank indicates not determined.

RESEARCH AND DEVELOPMENT, STABLE ISOTOPES

P. S. Baker F. R. Duncan

INTRODUCTION

Activities of the Research and Development Department continue to be divided between the service operations, by which special forms of stable isotopes are prepared in order to fill the specific needs of the eventual users, and the development projects, which are more intimately concerned with problems arising within the Stable Isotopes Research and Production Division.

SPECIAL SERVICES

As indicated previously,[1] the availability of special forms of enriched isotopes is an outgrowth of the increasing number of requests associated with the growing utilization of separated material. The importance of this type of service is evidenced by the fact that it is continually becoming more difficult to keep up with the requests; the backlog now amounts to about 40 targets. The user of these services compensates this laboratory for all work done.

Target Preparation

In the past, specimens of copper, nickel, and iron isotopes have been prepared by powder-metallurgy processes on a number of occasions. During this period, additional targets, 0.018 × 0.750 × 1.250 in., of all three elements have been prepared for Harvard University. Details of the procedure are outlined in a previous report.[2]

Enriched nickel foils, too thin to be prepared by powder metallurgy, are prepared by electroplating nickel from an $NiSO_4$ bath onto an aluminum strip, which subsequently is dissolved in dilute HCl, leaving the elemental nickel.[3] Targets, 0.003 in. thick and 2 cm in diameter, of both Ni^{58} and Ni^{60} have been prepared in this way for the Physics Department of the University of Minnesota.

Thin foils of Cu^{63} and Cu^{65} were electrodeposited for the first time in a similar manner. In this case, electrodeposition from cyanide solution has proved to be superior to deposition from a sulfate bath, and magnesium is more satisfactory as a base than is aluminum, since the latter is attacked by the highly alkaline cyanide bath.

About 30 g of B^{10} was converted to B_2O_3 by oxidation with concentrated HNO_3, recrystallization from water, and dehydration of the resulting H_3BO_3 by fusing it under a mild vacuum. By the same procedure, except for the omission of the final dehydration, about 10 g of elemental B^{10} was converted to H_3BO_3.

One- and two-gram samples, respectively, of Li^6 and Li^7 of very high chemical purity were prepared by repeated vacuum distillations. These samples are to be used by the University of Michigan in measurements of bulk properties. An Li^6 target was also prepared for use in the ORNL Van de Graaff accelerator.

Special Procedures

With every special service, unusual techniques and procedures have to be developed. One of these developments which has been evolved in the past six months follows:

Thin foils of Fe^{54} and Fe^{56} are currently being prepared, but with some difficulty. Normal commercial plating baths of the chloride or sulfate type have not deposited suitable thin foils of isotopic material. The chloride bath is hard to maintain in proper balance when the amount of available starting material is limited and when dragout and spray are excessive. Sulfate baths, on the other hand, yield deposits of a very brittle nature, which makes them extremely difficult to handle without breakage. A more satisfactory bath comprises an iron salt complexed with triethanolamine and the sodium salt of ethylenediaminetetraacetic acid.[4] But, as yet, no backing (which is suitable to the user and which will stand up under the conditions of plating) has been found. Backings of platinum or tantalum, which might ordinarily be considered, are entirely unsatisfactory from the standpoint of the user. Experiments are now being performed in which the iron is plated on copper and the copper is then ground away mechanically.

[1] P. S. Baker *et al.*, *Stable Isotope Research and Production Semiann. Prog. Rep. Nov. 20, 1954*, ORNL-1829, p 42.

[2] P. S. Baker, F. R. Duncan, and H. B. Greene, *Stable Isotope Research and Production Semiann. Prog. Rep. May 20, 1955*, ORNL-1908, p 42.

[3] P. S. Baker *et al.*, *Stable Isotope Research and Production Semiann. Prog. Rep. Nov. 20, 1954*, ORNL-1829, p 44.

[4] E. F. Foley, Jr., H. B. Linford, and W. R. Meyer, *Plating* 40, 887 (1953).

Cooperative Calutron-Cyclotron Program

The importance of separated stable isotopes as target materials for cyclotron-produced radioisotopes has been indicated,[3,5] and the necessity of fabricating Ca^{48} targets has been anticipated.

In order that Ca^{47} may be prepared efficiently and economically by cyclotron bombardment of Ca^{48}, it is desirable that a relatively thin layer of Ca^{48}, in the form of elemental calcium, be deposited on a standard silver target base. The problems encountered consist chiefly in converting the small available amount of enriched material (inventoried as $CaCO_3$) to the elemental form, depositing it as an adherent layer within the target area to be subjected to the cyclotron beam, and then protecting this layer from atmospheric attack during the pre- and postrun handling. In addition, unused portions of the starting material must be recoverable, and the irradiated calcium on the target must lend itself to a minimum of chemical processing.

These requirements have been met with the development of the following procedure: The $CaCO_3$ is calcined; then the CaO is reduced with powdered aluminum at 1200°C in a vacuum. The reduction is shown by the equation

$$6CaO + 2Al \longrightarrow 3Ca + Al_2O_3 \cdot 3CaO$$

The elemental calcium which is formed is simultaneously evaporated onto the cyclotron target. While still under vacuum, the calcium layer is covered with a thin layer of aluminum which is evaporated from a second vapor source. The aluminum layer protects the calcium from atmospheric oxidation without affecting seriously the cyclotron bombardment.

Figures 12*a* and 12*b* are photographs of the equipment, with the vacuum bell jar in place, and removed, respectively. The furnace for the reduction-evaporation of the calcium includes, in addition to a vapor-directing duct, a die-formed tantalum sheet, which is formed so as to provide a receptacle for pellets of an intimate mixture of CaO and aluminum. The furnace, clamped between water-cooled copper busbars, is visible in the lower center portion of the bell-jar base.

In the lower front portion may be seen a helical tungsten filament, which supports short sections of aluminum wire; this arrangement provides the final protective layer of aluminum. Located about 4 in. above these assemblies is the silver target, strung with a resistance heater of Nichrome wire.

During operations the bell jar is evacuated, the target is heated to 300°C, and the tantalum furnace is heated to about 1200°C by a current from a conventional arc-welding machine being passed directly through it. The reduction-evaporation is complete in a few minutes; the deposit is then flashed with aluminum from the tungsten filament. After being cooled, the target is ready for bombardment. Typical quantities deposited are 55 to 60 mg of calcium, and 10 to 15 mg of aluminum.

A number of satisfactory normal calcium targets have been prepared in this way. The final plating of the targets with enriched Ca^{48} is awaiting cyclotron scheduling. It is anticipated that this equipment and a similar procedure will be used for the preparation of Mg^{25} (for Na^{22} production) and

Fig. 12*a*. Apparatus for Vacuum-Plating Cyclotron Targets. Vacuum bell jar in place.

[5] P. S. Baker, F. R. Duncan, and H. B. Greene, *Stable Isotope Research and Production Semiann. Prog. Rep. May 20, 1955*, ORNL-1908, p 44.

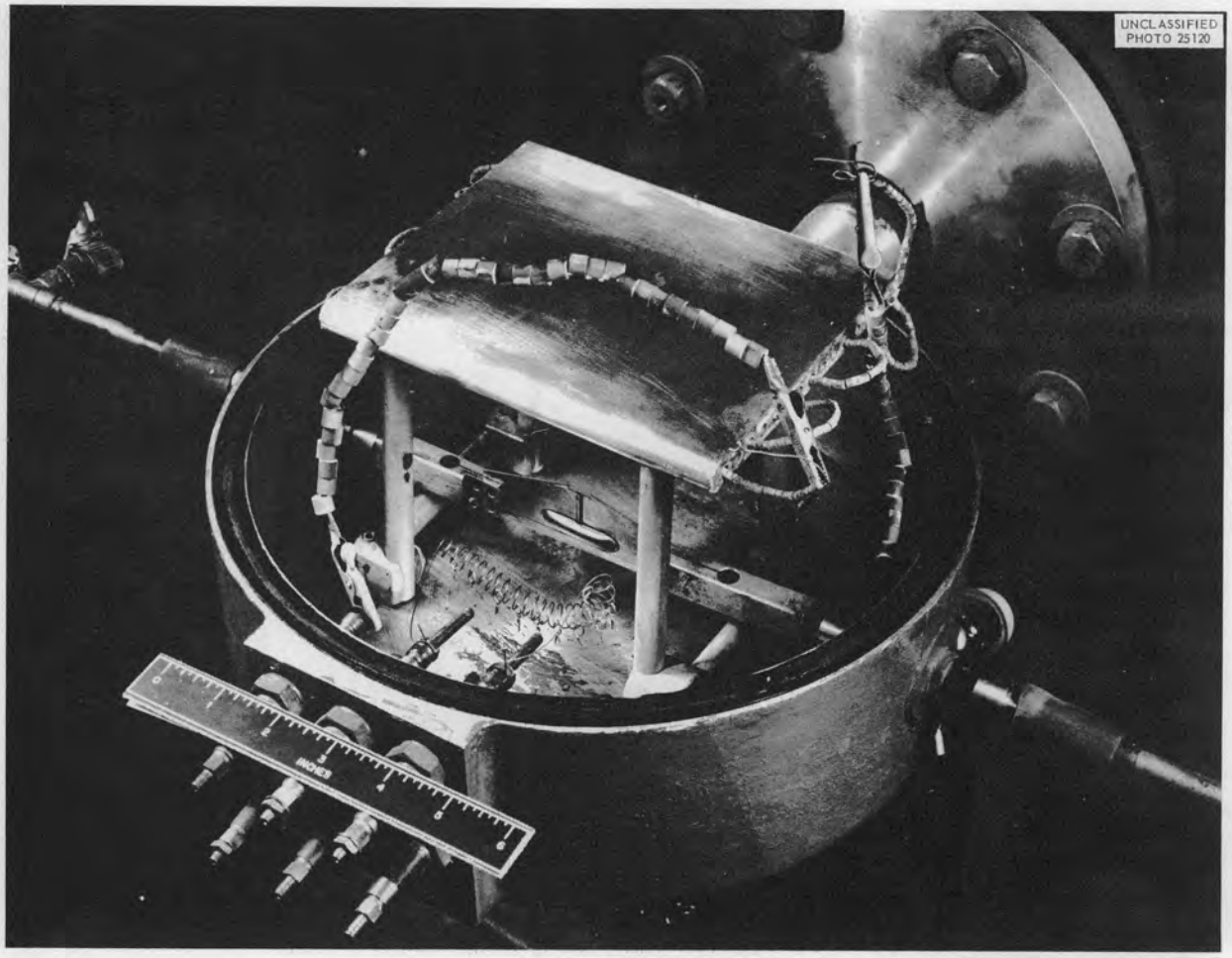

Fig. 12*b*. Apparatus for Vacuum-Plating Cyclotron Targets. Vacuum bell jar removed.

Mg^{26} (for Al^{26} production) targets, although the evaporation of aluminum should not be necessary.

Services for Production Department

Considerable difficulty was encountered in maintaining stable calutron operations during the separation of barium isotopes. It was suggested by the development group that the strontium content of the barium charge material might be the main cause of the trouble, since it had been found earlier[6,7] that strontium has an appreciably higher vapor pressure than barium at temperatures lower than the operating temperature for barium. To test the hypothesis, several batches of barium metal, consisting of approximately 200 g of barium each, were vacuum-distilled to reduce the strontium content from about 4 to less than 0.5%. The calutron operations improved considerably when this charge material was used.

From time to time, batches of samarium chloride and other powdery charge materials have been fused into solid masses to prevent air entrainment and subsequent blowouts during the evacuation of the calutron vacuum chamber.

RELATED WORK

Investigation of Zone Melting

An allied project has been the design and construction of an apparatus for the study of chemical

[6] P. S. Baker, F. R. Duncan, and H. B. Greene, *Stable Isotope Research and Production Semiann. Prog. Rep. Nov. 20, 1953*, ORNL-1617, p 17.

[7] L. Brewer, *Chemistry and Metallurgy of Miscellaneous Materials: Thermodynamics* (ed. by L. L. Quill), NNES IV-19B, p 31, McGraw-Hill, New York, 1950.

purifications by means of the zone-melting procedure. It has been suggested, for example, that the technique might be used for the separation of rare earths from each other. The availability of such equipment within the Division will undoubtedly lead to its use as a purification device in the preparation of spectrometric standards and in the purification of samples for physical and nuclear property studies. Isotopically enriched material such as lithium, barium, and calcium could be prepared, in all probability, with higher chemical purities than are now possible with vacuum distillation.

Figure 13 is a photograph of the equipment in its present state of development. It consists of a screw-driven carriage powered by an electric motor through a variable-speed transmission. The carriage may be advanced at any speed up to a maximum of 3 in./min.

Mounted on the carriage is a Transite rack capable of supporting a boat filled with the material to be purified. The boat is carried through the field of an induction heater coil for melting, although a resistance heater can also be used.

Some preliminary runs with alloys of lead and tin were carried out to check the mechanical operation. At present the apparatus is being adapted to the melting of rare-earth fluorides. The problems encountered are primarily caused by temperature and zone-length control factors and by the necessity for container materials which will not corrode appreciably when in contact with molten rare-earth fluorides at temperatures in excess of

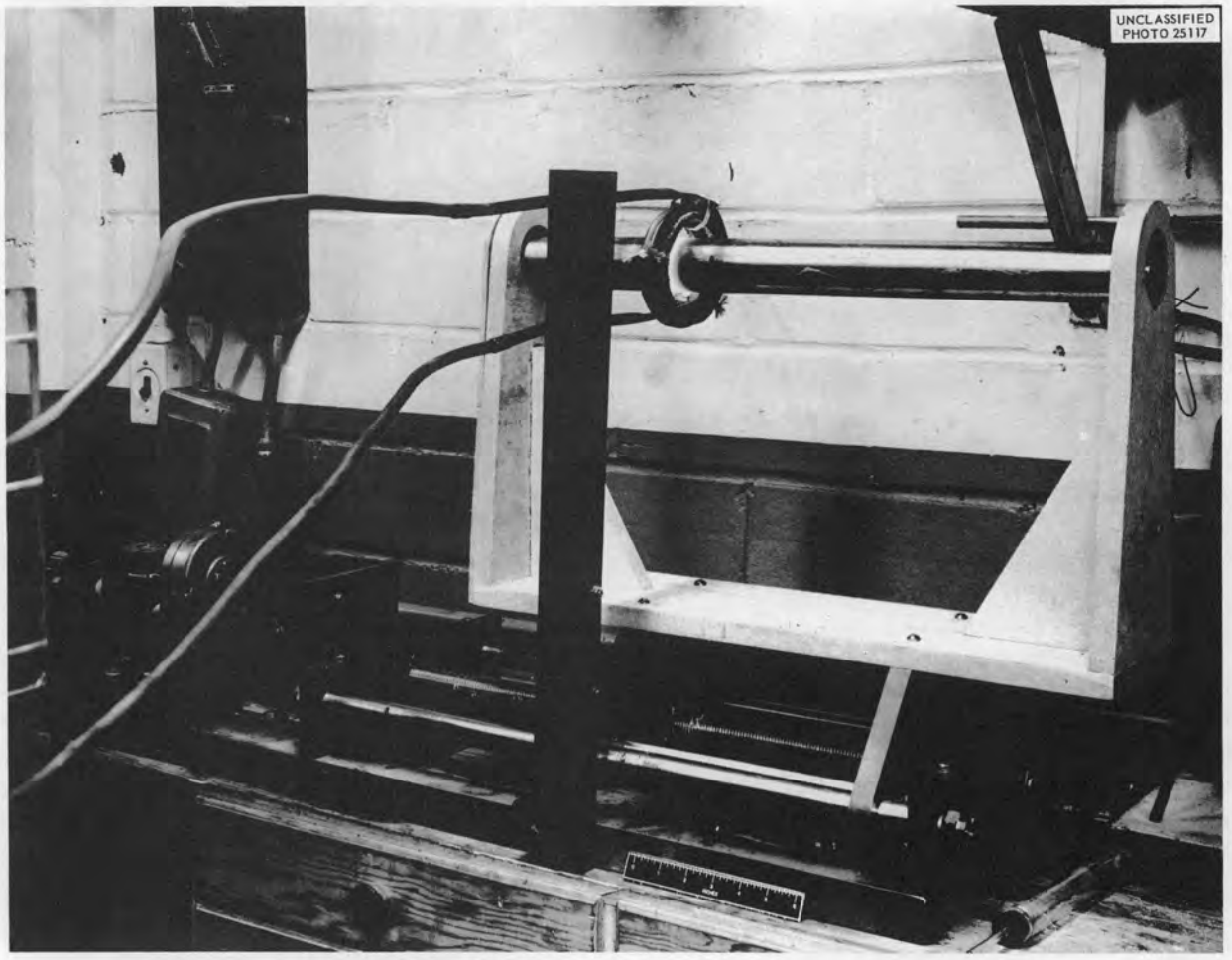

Fig. 13. Zone-Melting Apparatus.

1000°C. Platinum appears to be reasonably satisfactory, and a 10-in. boat has been fabricated for use in the actual zone-melting tests.

The length of the molten zone can be controlled to some extent by surrounding the boat with an Inconel sleeve. A section of this sleeve is heated by the induction coil, and then the platinum boat and contents are heated by radiation and conduction. A thermocouple between the Inconel sleeve and platinum boat automatically controls the temperature of both by turning the induction heater off and on.

Radioisotope Separation Study

During this period the need for, and feasibility of, separating radioactive isotopes was investigated in a preliminary manner. It is apparent that a demand exists both for carrier-free radioisotopes and for radioisotopes of very high specific activity for use in high-resolution and compact teletherapy and radiography sources. There is also a need for high specific activities for the fundamental studies of the nuclear properties of the nuclides themselves.

While a number of mildly active isotopes, including U^{235}, U^{236}, Rb^{87}, Re^{187}, Sn^{124}, Sm^{147}, K^{40}, and Be^{10}, have already been separated readily with calutron equipment, there still remain others which could be separated without serious modifications to existing equipment. For high-energy gamma emitters, however, new facilities for separation and handling are mandatory. It is in this class that most of the desired isotopes are found, including Co^{60}, Cs^{134}, Sr^{89}, and Sr^{90}.

It has been suggested that electromagnetic source equipment for this type of separation be made up of functional subunits, so arranged that any component that fails in service can be removed and replaced by remote control without the necessity for complete chemical and mechanical servicing of the entire unit. It has also been suggested that the receiver be designed so that individual collectors can be readily handled through recovery chemistry.

Remote handling of all systems behind suitable barriers appears to be practical. Wash chemistry and recovery chemistry would be handled in areas isolated from operational and servicing areas.

Laboratory Expansion

The accumulation of equipment used in the various target fabrications had overcrowded existing laboratory space. More bench area was needed which would be suitable for chemical treatment, as well as additional space for storing apparatus components. The laboratory space was essentially doubled by adding to the laboratory a former change room. Figure 14 shows the arrangement of the equipment. A 100-ton press, designed and fabricated specifically for isotope powder metallurgy, can be seen. Also visible is a hand-operated rolling mill for foil fabrication.

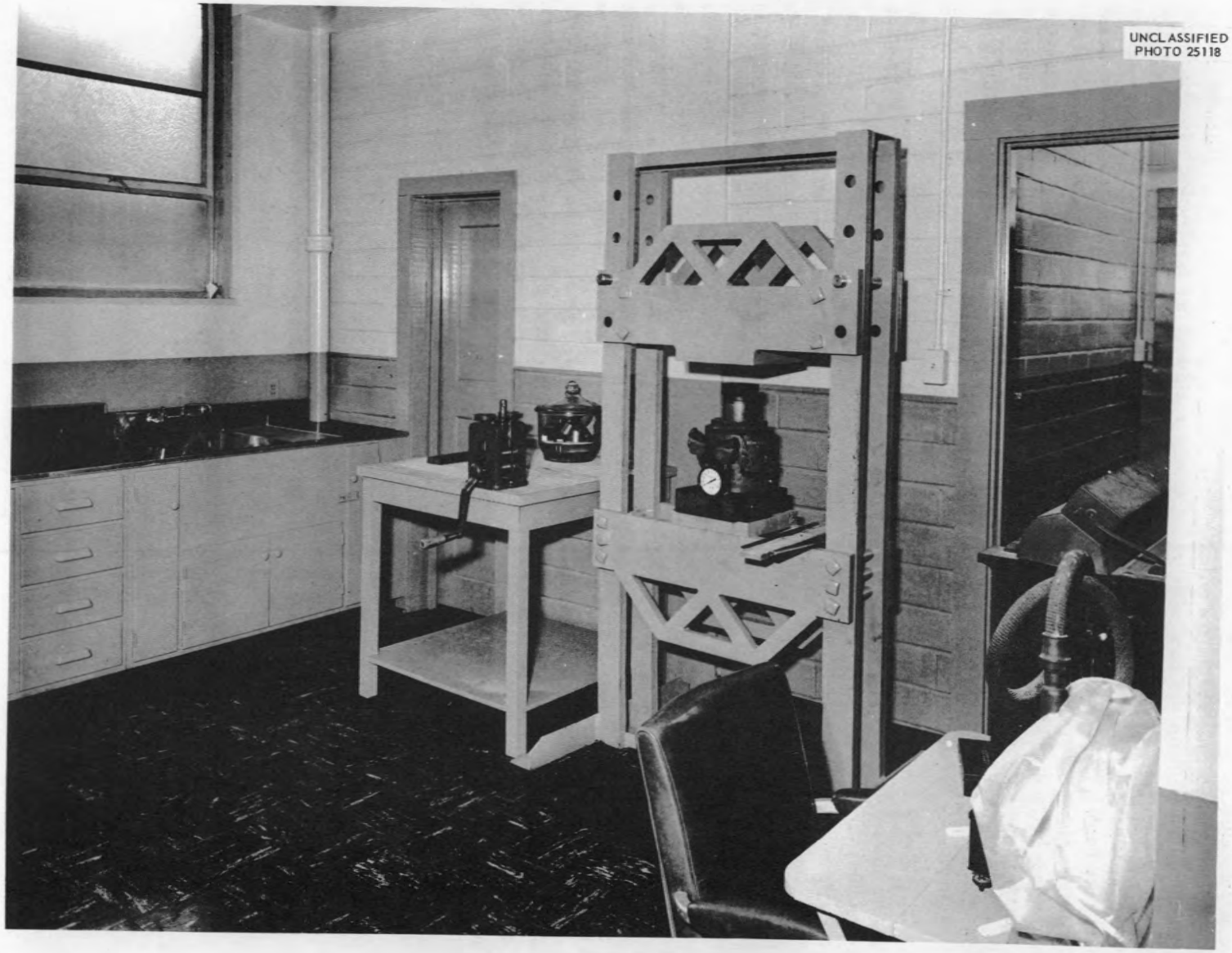

Fig. 14. Expanded Laboratory Facilities.

UTILIZATION OF ENRICHED STABLE ISOTOPES

The shipments of enriched stable isotopes that were made during the ten-year period (1946 through November 20, 1955) are illustrated in Fig. 15. It will be noted that the shipments through CY-1955 have already exceeded the total for the best previous year by about 5%. This can be ascribed to an increase in foreign sales and to the availability of special target forms of the isotopes. It is also noteworthy that, for the past few years, the percentage of on-project users has decreased. For the seven years through early 1953, about 65% of the total shipments were on-project; during 1953 the number of on-project shipments made up 45% of the total; during 1954, 48%; and during 1955 (through November), only 44%.

Table 20 gives a summary of the isotope shipments during the period from May 20, 1955, through November 20, 1955. The asterisks in this table indicate those shipments which required special services (see section "Research and Development"). About 13% of the total falls in this category. Table 21 indicates the distribution of the shipments as well as the number of shipments by months.

Table 22 is a list of isotopes which have been allocated but have not yet been shipped. Table 23 is a list of pending requests for isotopes which have not been approved by the AEC. Included are the reasons for withholding approval.

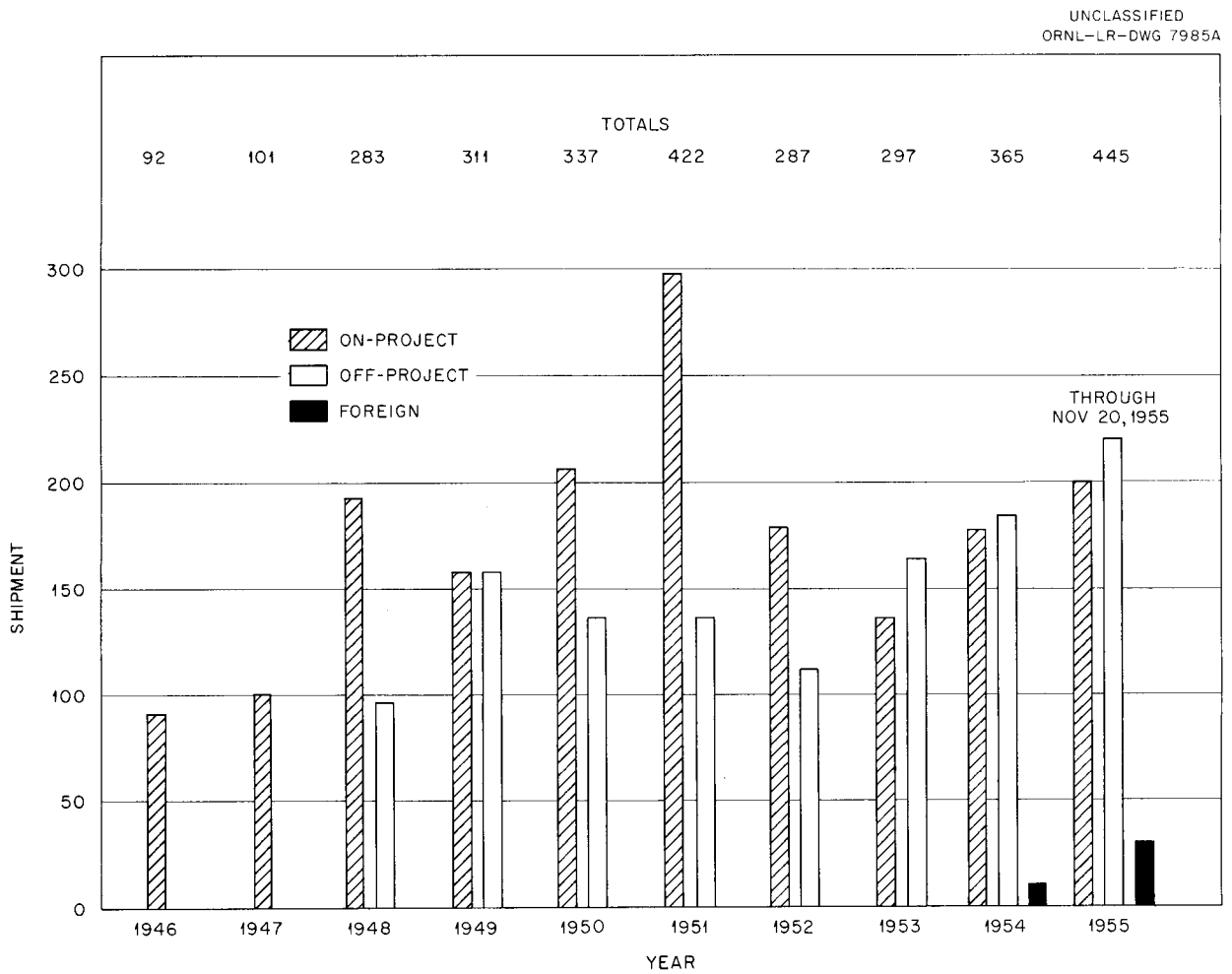

Fig. 15. Shipments of Enriched Stable Isotopes – 1946 to 1955.

TABLE 20. SUMMARY OF ISOTOPE SHIPMENTS, MAY 20, 1955 THROUGH NOVEMBER 20, 1955

Request No.	Requester	Isotope	Lot	Quantity (mg)
B-5113-S	Oak Ridge National Laboratory	Ru^{104}	GJ 796(b)	75
B-5126-S	Oak Ridge National Laboratory	Ru^{99}	GJ 792(a)	75
B-5128-S	Oak Ridge National Laboratory	Ru^{101}	GJ 794(a)	150
B-5514	Princeton University	Si^{30}	EB 526(a)	50
B-5516	Princeton University	Mg^{25}	DZ 520(a)	50
B-5554-S	Argonne National Laboratory	Ru^{102}	GJ 795(b)	5
B-5569	Carnegie Institution of Washington	K^{41}	EY 626(e)	30
B-5576	Case Institute of Technology	Li^{6}	FY 745(be)	6,300
B-5577	University of Pennsylvania	Hf^{180}	GP 830(a)	2
B-5545	Carnegie Institution of Washington	Ru^{99}	GJ 792(a)	50
B-5546	Carnegie Institution of Washington	Ru^{101}	GJ 794(a)	100
B-5468	Stanford University	Mg^{24}	P 47(a)	500
B-5594-S	Brookhaven National Laboratory	Ni^{58}	GT 844(a)	15,000
F-2365-T	Centre National de la Recherche Scientifique (CNRS), New York Office (France)	S^{34}	ES 598(a)	300
B-4218-S	Argonne National Laboratory	Ru^{104}	GJ 796(b)	10
B-5102-S	Brookhaven National Laboratory	Hf^{178}	GP 828(a)	3,800
B-5141*	Westinghouse Research Laboratory	Ni^{58}	FJ 669(a)	21,241
B-5159*	Westinghouse Research Laboratory	Cu^{63}	GT 849(a)	32,858
B-5160*	Westinghouse Research Laboratory	Cu^{65}	GT 850(a)	9,957
B-5363-S	Argonne National Laboratory	Ru^{104}	GJ 796(b)	10
B-5513	Princeton University	S^{33}	GB 758(a)	35
B-5515	Princeton University	Si^{29}	GC 762(a)	50
B-5646	Yale University, Sloane Laboratory	Mg^{24}	P 47(a)	500
B-5547	Carnegie Institution of Washington	Zn^{64}	EK 556(a)	150
B-5628-S	Brookhaven National Laboratory	Sb^{121}	AE 93(ar)	15
B-5659-S	Oak Ridge National Laboratory (Ketelle)	Ce^{138}	FR 707(a)	100
F-2419-T	CNRS, New York Office (France)	Hg^{198}	DR 486(d)	10
F-2420-T	CNRS, New York Office (France)	Hg^{200}	DR 488(e)	10
F-2421-T	CNRS, New York Office (France)	Hg^{202}	DR 490(g)	10
F-2422-T	CNRS, New York Office (France)	Hg^{204}	DR 491(e)	10
B-4972	Carnegie Institution of Washington	Ru^{104}	GJ 796(a)	200
B-5456	California Institute of Technology	Zr^{90}	CK 334(ar)	50
B-5457	California Institute of Technology	Zr^{94}	EE 542(a)	50
B-5588	Bartol Research Foundation	Sn^{124}	FS 719(a)	100
B-5641-S	Argonne National Laboratory	Mo^{100}	AW 169(a)	10

*Special services.

TABLE 20 (continued)

Request No.	Requester	Isotope	Lot	Quantity (mg)
B-5642-S	Argonne National Laboratory	Zr^{96}	GQ 835(a)	25
B-5647	Columbia University	Ba^{132}	FU 728(a)	20
B-5648	Columbia University	Ca^{42}	FF 649(a)	10
F-2435-T	Australian Scientific Liaison	S^{33}	CX 392(b)	10
B-5682-S	Los Alamos Scientific Laboratory	Cd^{112}	CE 318(a)	20
B-5690*	Carnegie Institution of Washington	Ca^{43}	FO 691(a)	50
B-5700-S	Brookhaven National Laboratory	Cd^{113}	AB 86(a)	50
B-5701-S	Brookhaven National Laboratory	In^{115}	CO 355(a)	100
B-5680-S	Oak Ridge National Laboratory	Ce^{136}	FR 706(a)	100
B-5724-S	Oak Ridge National Laboratory	Ca^{48}	GV 860(a)	5
B-5632-S	Argonne National Laboratory	Ca^{48}	GV 860(b)	10
B-5679-S	Oak Ridge National Laboratory (Moak)	Li^{6}	HD 885(a)	50
B-5691	Carnegie Institution of Washington	Te^{122}	DX 506(a)	50
B-5692	Carnegie Institution of Washington	Te^{128}	FR 646(a)	100
B-5737	University of Wisconsin	Cd^{111}	AC 89(a)	50
F-2428-T	CNRS, Air Cargo (France)	Li^{6}	HD 885(a)	300
B-5414-S	Brookhaven National Laboratory	Li^{7}	FI 668(i)	10,000
B-5605	University of Arkansas	Sr^{86}	FL 680(a)	50
B-5606	University of Arkansas	Sr^{88}	EH 551(a)	40
B-4927	Ohio State University	Ru^{98}	GJ 791(a)	50
B-4928	Ohio State University	Ru^{99}	GJ 792(a)	50
B-4930	Ohio State University	Ru^{101}	GJ 794(a)	100
B-5643	Ohio State University	Gd^{158}	GK 803(a)	100
B-5644	Ohio State University	Gd^{160}	GK 804(a)	150
B-5676	Stanford University	B^{11}	GF 769(a)	250
B-5677-S	University of California Radiation Laboratory	Pb^{204}	EO 581(a)	500
B-5684-S	Brookhaven National Laboratory	Se^{74}	DY 513(a)	230
B-5685-S	Brookhaven National Laboratory	Se^{76}	FG 655(a)	375
B-5686-S	Brookhaven National Laboratory	Se^{77}	BS 270(ar)	500
B-5687-S	Brookhaven National Laboratory	Se^{78}	CY 400(ar)	1,030
B-5688-S	Brookhaven National Laboratory	Se^{80}	BS 272(a)	820
B-5689-S	Brookhaven National Laboratory	Se^{82}	DY 518(a)	1,080
B-5694	Columbia University	Pb^{204}	EO 581(a)	15
B-5705	Naval Research Laboratory	Br^{79}	EJ 552(a)	91
B-5718	Carnegie Institution of Washington	Sr^{86}	EH 549(a)	25

*Special services.

TABLE 20 (continued)

Request No.	Requester	Isotope	Lot	Quantity (mg)
B-5759-S	Oak Ridge National Laboratory (shipment to Canada)	La^{138}	GO 823(a)	750
B-5730	University of Washington, Seattle	Ni^{58}	GT 844(a)	100
B-5731	University of Washington, Seattle	Cu^{63}	FJ 673(a)	100
B-5740-S	Brookhaven National Laboratory	Ru^{99}	GJ 790(b)	40
B-5741-S	Brookhaven National Laboratory	Ru^{99}	GJ 792(a)	100
F-2468-T	Federal Republic of Germany	Hg^{202}	DR 490(a)	3
B-5760	Ohio State University	Hg^{204}	DR 491(hs)	145
B-5127-S	Oak Ridge National Laboratory	Ru^{100}	GJ 793(a)	150
B-5744	Columbia University	K^{40}	GN 821(a)	3
B-5829-S	University of California (LASL)	S^{34}	GB 759(a)	500
B-5125-S	Oak Ridge National Laboratory	Ru^{98}	GJ 791(a)	70
B-5518	Yale University	B^{10}	SS 1(ad)	30,000
B-5772	University of Michigan	Ti^{46}	EN 576(a)	150
B-5742	University of Wisconsin	Mo^{95}	FP 697(b)	10
B-5743	University of Wisconsin	Mo^{97}	FP 699(b)	20
B-5679-S	Oak Ridge National Laboratory	Li^{6}	I 30-33(c)	50
B-5868-S	Oak Ridge National Laboratory	Li^{7}	FI 668(j)	3,641
B-5869-S	Oak Ridge National Laboratory	Li^{6}	FY 745(bd)	2,957
B-5863-S	Brookhaven National Laboratory	Cd^{114}	CE 320(a)	40
F-2572-T	Atomic Energy of Canada, Ltd	Pb^{207}	EO 583(a)	40
F-2573-T	Atomic Energy of Canada, Ltd	Pb^{208}	EO 584(a)	40
B-5821	University of Wisconsin	Se^{78}	FG 657(d)	100
B-5822	University of Wisconsin	Se^{80}	FG 658(b)	100
B-3780	Johns Hopkins University	Rb^{85}	GX 862(a)	700
B-5804-S	Oak Ridge National Laboratory	Tl^{203}	CH 326(br)	250
B-5805-S	Oak Ridge National Laboratory	Pt^{194}	GH 778(a)	300
B-5806-S	Oak Ridge National Laboratory	Tl^{205}	BV 279(a)	500
B-5807-S	Oak Ridge National Laboratory	Pb^{204}	EO 581(a)	500
B-5847	University of Michigan	W^{184}	CL 343(ar)	50
B-5848	University of Michigan	W^{180}	EL 561(a)	20
B-5900	University of Michigan	Li^{7}	FI 668(j)	3,000
B-5786	Carnegie Institution of Washington	Te^{124}	CA 295(a)	20
B-5787	Carnegie Institution of Washington	Te^{126}	CA 297(a)	100
B-5838	Naval Research Laboratory	Ni^{61}	GT 846(a)	120
B-5839	Naval Research Laboratory	V^{50}	FM 683(br)	25
B-5851	Naval Research Laboratory	K^{41}	EY 626(e)	100

TABLE 20 (continued)

Request No.	Requester	Isotope	Lot	Quantity (mg)
B-5887	Harvard University	Pb^{208}	EO 584(a)	915
B-5890	Harvard University	Fe^{56}	DO 478(ar)	1,800
B-5790	California Institute of Technology	W^{186}	CL 344(ar)	25
B-5791	California Institute of Technology	W^{180}	CL 340(ar)	5
B-5792	California Institute of Technology	Hf^{174}	GP 825(a)	25
B-5824	The Firestone Tire and Rubber Company	Li^{7}	FI 668(i)	1,000
F-2594-T	CNRS, New York Office (France)	Hg^{198}	DR 486(b)	10
F-2595-T	CNRS, New York Office (France)	Hg^{199}	DR 487(as)	10
F-2596-T	CNRS, New York Office (France)	Hg^{200}	DR 488(a)	10
F-2597-T	CNRS, New York Office (France)	Hg^{201}	DR 489(a)	10
F-2598-T	CNRS, New York Office (France)	Hg^{202}	DR 490(a)	10
F-2599-T	CNRS, New York Office (France)	Hg^{204}	DR 491(b)	10
B-5904-S	Oak Ridge National Laboratory	Pb^{207}	EO 583(a)	365
B-5876	Esso Research and Engineering Co.	V^{50}	FM 683(b)	10
B-5877	Esso Research and Engineering Co.	Fe^{54}	FN 685(b)	10
B-5878	Esso Research and Engineering Co.	Ni^{62}	FJ 672(a)	10
B-5858	University of California	Hf^{177}	GP 827(a)	500
B-5859	University of California	Hf^{179}	GP 829(b)	150
B-5922-S	Knolls Atomic Power Laboratory	Ta^{180}	GY 864(c)	50
No Number	Brookhaven National Laboratory	Li^{6}	SS 5(a)	10,300
B-4812	Columbia University	Rb^{87}	GX 863(a)	30
B-5265	Carnegie Institution of Washington	Rb^{87}	GX 863(a)	50
B-5880-S	University of California Radiation Laboratory	Pd^{110}	GG 775(a)	500
B-5889	Harvard University	Ni^{58}	GT 844(a)	1,850
B-5926	Stanford University	B^{10}	SS 1(a)	250
B-5927	Stanford University	Zn^{64}	EK 556(a)	959
B-5940-S	Argonne National Laboratory	Te^{130}	CA 299(a)	1,000
B-5757	Princeton University	Rb^{87}	GX 863(a)-2	500
B-5979-S	University of California (LASL)	Se^{80}	FG 658(a)	200
B-5980-S	University of California (LASL)	Se^{82}	FG 659(a)	160
B-5981-S	University of California (LASL)	Se^{82}	FG 659(b)	40
F-2630-T	Federal Republic of Germany	Pt^{190}	GH 776(a)	1
F-2631-T	Federal Republic of Germany	Pt^{192}	GH 777(a)	1
B-5998-S	Oak Ridge National Laboratory	Li^{6}	SS 5(a)	23,000
B-4113	Carnegie Institution of Washington	V^{50}	FM 683(b)	20

TABLE 20 (continued)

Request No.	Requester	Isotope	Lot	Quantity (mg)
B-5400	Johns Hopkins University	Li^6	SS 5(a)	2,000
B-5905-S	Oak Ridge National Laboratory	Ag^{107}	SU 41(br)	200
B-5971	Florida State University	Ge^{70}	BD-BE 205-210(a)	700
B-6037-S	Oak Ridge National Laboratory	Sn^{120}	EC 534(a)	25
B-6031-S	Oak Ridge National Laboratory	Cr^{50}	GS 840(a)	100
B-5946	U.S. Naval Radiological Defense Laboratory	W^{180}	EL 561(a)	16
B-5989	University of Wisconsin	Sm^{144}	GM 813(b)	250
B-6007-S	Brookhaven National Laboratory	B^{11}	GF 769(a)	200
B-6010-S	Argonne National Laboratory	Se^{82}	BS 273(ar)	500
B-6011-S	Argonne National Laboratory	Sr^{86}	FL 680(a)	500
B-6013-S	Argonne National Laboratory	Rb^{85}	GX 862(a)	500
B-6014-S	Argonne National Laboratory	Rb^{87}	GX 863(a)-3	500
B-6015-S	Argonne National Laboratory	Sr^{88}	FL 682(a)	2,000
B-6016-S	Argonne National Laboratory	Se^{76}	CY 398(ar)	250
B-6047	University of Michigan	W^{186}	CL 344(a)	100
B-4504*	Duke University	Fe^{54}	DO 477(a)	30,000
None	Oak Ridge National Laboratory	Cr^{54}	GR 839(c)	140
B-5519	Michigan State University	Li^6	SS 5(a)	1,045
B-5520	Michigan State University	Li^7	FI 668(a)	1,028
B-5908	University of Minnesota	Cu^{65}	DU 498(a)	150
B-5909	University of Minnesota	Cu^{63}	FJ 673(a)	301
B-5912	University of Minnesota	Ni^{58}	GT 844(a)	233
B-5913	University of Minnesota	Ni^{60}	FJ 670(a)	278
B-6056-S	University of California Radiation Laboratory	Cd^{106}	AC 88(a)	50
B-5974-S	University of California Radiation Laboratory	Mo^{98}	RT 58(a)	100
B-6045-S	Los Alamos Scientific Laboratory	Cr^{50}	GZ 868(a)	1,390
B-6065-S	Los Alamos Scientific Laboratory	Hf^{176}	GP 826(a)	200
B-6066-S	Los Alamos Scientific Laboratory	Hf^{177}	GP 827(a)	200
B-6067-S	Los Alamos Scientific Laboratory	Hf^{178}	GP 826(b)	200
B-6068-S	Los Alamos Scientific Laboratory	Hf^{179}	GP 829(a)	200
B-6069-S	Los Alamos Scientific Laboratory	Hf^{180}	GP 830(a)	200
B-6071	Indiana University	Te^{126}	CA 297(ar)	500
B-6076-S	Phillips Petroleum Co.	Rb^{87}	GX 863(a)	800
B-6077-S	Phillips Petroleum Co.	Sb^{123}	AE 94(ar)	262

*Special services.

TABLE 20 (continued)

Request No.	Requester	Isotope	Lot	Quantity (mg)
B-6078-S	Phillips Petroleum Co.	Te^{123}	HC 877(b)	125
B-6102-S	Oak Ridge National Laboratory	Zr^{90}	EE 539(a)	30
F-2649-T	CNRS, New York Office (France)	B^{10}	SS 1(a)	10,000
F-2740-T	Atomic Energy of Canada, Ltd.	Sm^{152}	FH 665(ar)	10
B-6046	University of Chicago	Gd^{155}	GK 800(a)	25
B-6049	University of Chicago	Gd^{157}	GK 802(a)	25
B-6060	University of Chicago	Ti^{49}	GA 755(a)	20
B-6061	University of Chicago	Ti^{47}	BF 216(ar)	20
B-6063	University of Chicago	Ni^{61}	GT 846(a)	20
B-6080	Bartol Research Foundation	Re^{187}	CP 357(a)	50
B-6081	Bartol Research Foundation	Re^{185}	CP 356(a)	50
B-6089	University of Illinois	Pb^{204}	FQ 702(a)	500
B-6090	University of Illinois	Pb^{207}	FQ 704(a)	2,000
B-6091	University of Illinois	Pb^{206}	BO 258(ar)	500
B-6092	University of Illinois	Pb^{204}	EO 581(a)	500
B-6098	Purdue University	S^{36}	ES 599(a)	5
B-6099	Purdue University	Ca^{48}	GV 860(b)	10
B-6107-S	Brookhaven National Laboratory	Cr^{52}	GR 837(a)	2,000
B-6120-S	Oak Ridge National Laboratory	Sb^{121}	AE 93(ar)	30
B-6121	Oak Ridge National Laboratory	Sb^{123}	BT 275(a)	40

TABLE 21. DISTRIBUTION OF ISOTOPE SHIPMENTS FROM MAY 20, 1955, THROUGH NOVEMBER 20, 1955

Month	On Project	Off Project	Foreign	Total	Special Services
May	6	5		11	
June	11	17	6	34	4
July	16	16	2	34	1
August	10	8	2	20	2
September	8	20	6	34	7
October	16	13	2	31	4
November	18	20	2	40	7
Total	85	99	20	204	25

TABLE 22. ISOTOPES APPROVED BY AEC BUT NOT YET SHIPPED AS OF NOVEMBER 20, 1955

Authorization No.	Applicant	Isotope	Quantity (mg)	Sample Number
B-5142	Westinghouse Research Laboratory	Ge^{70}	10,200	FZ 747(a)
B-5143	Westinghouse Research Laboratory	Ge^{72}	15,500	FZ 748(a)
B-5144	Westinghouse Research Laboratory	Ge^{73}	3,150	FZ 749(a)
B-5145	Westinghouse Research Laboratory	Ge^{74}	17,600	FZ 750(a)
B-5146	Westinghouse Research Laboratory	Ge^{76}	4,800	FZ 751(a)
B-5147	Westinghouse Research Laboratory	W^{182}	8,000	EL 562(a)
B-5148	Westinghouse Research Laboratory	W^{183}	3,160	EL 563(a)
B-5149	Westinghouse Research Laboratory	W^{184}	8,470	EL 564(a)
B-5150	Westinghouse Research Laboratory	W^{186}	7,250	EL 565(a)
B-5151	Westinghouse Research Laboratory	Cr^{52}	23,100	EU 610(a)
B-5152	Westinghouse Research Laboratory	Zn^{66}	5,000	EK 557(a)
B-5153	Westinghouse Research Laboratory	Zn^{68}	2,920	EK 559(a)
B-5154	Westinghouse Research Laboratory	Ti^{46}	4,100	GA 752(a)
B-5155	Westinghouse Research Laboratory	Ti^{47}	2,620	GA 753(a)
			2,220	GA 753(b)
B-5156	Westinghouse Research Laboratory	Ti^{48}	35,600	GA 754(a)
B-5157	Westinghouse Research Laboratory	Ti^{49}	4,000	GA 755(b)
B-5158	Westinghouse Research Laboratory	Ti^{50}	1,130	GA 756(a)
			1,340	GA 756(b)
B-5218	The Rice Institute	Nd^{142}	200	EQ 587(ar)
B-5357	Westinghouse Research Laboratory	Cr^{50}	4,805	GS 840(a)
B-5439	Washington University	Tl^{203}	300	CC 307(a)
B-5467	Monsanto Chemical Company	Ca^{43}	15	FO 691(a)
B-5371	Wesleyan University	Zr^{91}	825	CV 382(a)
B-5373	Wesleyan University	Zr^{92}	1,000	CV 383(a)
B-5549	University of Kentucky	Ca^{43}	5	FO 691(a)
B-5663	Columbia University	Cr^{50}	1,000	GR 836(a)
B-4285	University of Arkansas	Ni^{58}	3,000	GT 844(a)
B-4929	Ohio State University	Ru^{100}	100	GJ 793(a)
B-5793	University of Connecticut	Sn^{116}	2,000	EC 530(a)
B-5794	University of Connecticut	Sn^{120}	2,000	FS 717(a)
B-5795	University of Connecticut	Sn^{122}	1,500	EC 535(a)
B-5797	Ohio State University	Ru^{104}	15	GJ 796(a)
B-5816	University of Michigan	Fe^{54}	200	FN 685(b)
B-5817	University of Michigan	Ni^{64}	200	GT 848(a)

TABLE 22 (continued)

Authorization No.	Applicant	Isotope	Quantity (mg)	Sample Number
B-5818	University of Michigan	Zr^{96}	100	GQ 835(a)
B-5829-S	Los Alamos Scientific Laboratory	S^{34}	500	GB 759(a)
B-5841	University of California	W^{183}	100	CL 342(a)
B-5910	University of Minnesota	Fe^{54}	200	FN 685(a)
B-5911	University of Minnesota	Fe^{56}	200	DO 478(a)
B-4470	Duke University	Rb^{87}	3,000	GX 863(a)
B-5107	University of Chicago	Rb^{87}	20	GX 863(a)
B-4614	Massachusetts Institute of Technology	Ca^{48}	35	FF 653(a)
B-5310	Columbia University	Te^{125}	3,000	HC 879(b)
B-5598-S	Phillips Petroleum Company	Mo^{95}	405	RT 55(a)
B-5602-S	Phillips Petroleum Company	W^{183}	350	CL 342(a)
B-5603-S	Phillips Petroleum Company	Hg^{199}	240	DR 487(ds)
B-5802	University of Kentucky	Ca^{48}	20	GV 860(a)
B-5994	State University of Iowa	Li^{6}	250	SS-5(a)
B-6085-S	Knolls Atomic Power Laboratory	Li^{7}	900	CU 379(a)
B-6086-S	Knolls Atomic Power Laboratory	Li^{7}	900	GE 767(l)
B-6087-S	Knolls Atomic Power Laboratory	Li^{7}	900	FI 668(j)
B-6062	University of Chicago	Mo^{95}	20	AW 165(a)
B-6070	Indiana University	Sn^{124}	300	FS 719(a)
B-6132	U. S. Naval Radiological Defense Laboratory	Sm^{144}	50	GM 813(b)
B-5460	Westinghouse Research Laboratory	Cr^{54}	4,000	GR 839(c)
B-5372	Wesleyan University	Zr^{90}	1,000	CK 334(ar)

TABLE 23. STABLE ISOTOPE REQUESTS PENDING APPROVAL BY AEC

Allocation No.	Applicant	Isotope	Amount (mg)	Reason for Withholding Approval	Status in Production Schedule
B-4489	Duke University	Ba^{134}	2,000	Awaiting chemical refinement of sample	Estimated completion, March 1956
B-4487	Duke University	Ba^{136}	6,000		
B-4488	Duke University	Ba^{137}	6,000		
B-4491	Duke University	Ca^{42}	2,000–4,000	Awaiting chemical refinement of sample	Estimated completion, December 1955
B-4493	Duke University	Ca^{43}	1,000		
B-3897	Yale University	Ca^{48}	100		
B-4310	Washington University	Ca^{48}	100–1,000		
B-4529	University of Michigan	Ca^{48}	500		
B-4540-S	Argonne National Laboratory	Ca^{48}	50		
B-4779	University of California	Ca^{48}	500–1,000		
B-5236-S	Brookhaven National Laboratory	Dy^{161}	2,000	Not now available	
B-5237-S	Brookhaven National Laboratory	Dy^{162}	2,000		
B-5238-S	Brookhaven National Laboratory	Dy^{163}	2,000		
B-5239-S	Brookhaven National Laboratory	Dy^{164}	2,000		
B-5626-S	Brookhaven National Laboratory	Eu^{151}	20	Awaiting chemical refinement of sample	Estimated completion, April 1956
B-5627-S	Brookhaven National Laboratory	Eu^{153}	20		
B-6004-S	Brookhaven National Laboratory	Ge^{70}	500–1,000	Availability of quantity	
B-5248-S	Argonne National Laboratory	Ge^{73}	700	Availability of quantity	
B-4553	Carnegie Institute of Technology	Li^{6}	50,000	Domestic sales information	
B-5032	Massachusetts Institute of Technology	Mg^{25}	90	Availability of quantity	
B-5561-S	Oak Ridge National Laboratory	Mo^{100}	40	Availability of quantity	
B-5888	Harvard University	Ni^{60}	2,000–2,500	Additional information from applicant	
B-5567-S	Oak Ridge National Laboratory	Pd^{102}	50–150	Availability of quantity	

TABLE 23 (continued)

Allocation No.	Applicant	Isotope	Amount (mg)	Reason for Withholding Approval	Status in Production Schedule
B-5778	Carnegie Institution of Washington	K^{40}	5–50	Availability of quantity	
B-5837	Naval Research Laboratory	K^{40}	10	Availability of quantity	
B-5902-S	Oak Ridge National Laboratory	Re^{187}	300	Availability of quantity	
B-5664	Columbia University	Sm^{150}	910	Additional information from applicant	
B-5607	University of Arkansas	Sr^{84}	50	Awaiting chemical refinement of sample	Estimated completion, December 1955
B-4495	Duke University	Sr^{86}	4,000		
B-5605	University of Arkansas	Sr^{86}	50		
B-4496	Duke University	Sr^{87}	4,000		
B-5250-S	Argonne National Laboratory	Sr^{87}	700		
B-5606	University of Arkansas	Sr^{88}	50		
B-4603	Duke University	S^{33}	200	Availability of quantity and enrichment	
B-5308	Columbia University	Te^{123}	3,000	Availability of quantity	
B-4469	Duke University	Tl^{203}	5,000–10,000	Availability of quantity	
B-5461	Westinghouse Research Laboratory	Ti^{49}	1,640	Availability of quantity	
B-5662	Columbia University	Ti^{50}	1,000	Availability of quantity	
B-5711-S	Ames Laboratory	W^{182}	1,000–1,500	Availability of quantity	
B-5422	Naval Research Laboratory	W^{183}	3,100	Availability of quantity	
B-5712-S	Ames Laboratory	W^{183}	1,000–1,500	Availability of quantity	
B-5713-S	Ames Laboratory	W^{184}	1,000–1,500	Availability of quantity	
B-6001	Bartol Research Foundation	W^{184}	500	Availability of quantity	
B-5424	Naval Research Laboratory	W^{186}	5,000	Availability of quantity	
B-5714-S	Ames Laboratory	W^{186}	1,000–1,500	Availability of quantity	