UNCLASSIFIED

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES
3 4456 0350302 8

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION

ORNL-2114
Progress

cy. 5

# STABLE ISOTOPES DIVISION

## SEMIANNUAL PROGRESS REPORT

## FOR PERIOD ENDING MAY 20, 1956

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION

LIBRARY LOAN COPY

DO NOT TRANSFER TO ANOTHER PERSON

If you wish someone else to see this document, send in name with document and the library will arrange a loan.

OAK RIDGE NATIONAL LABORATORY
OPERATED BY
UNION CARBIDE NUCLEAR COMPANY
A Division of Union Carbide and Carbon Corporation
UCC
POST OFFICE BOX P · OAK RIDGE, TENNESSEE

UNCLASSIFIED

Printed in USA. Price 40 cents. Available from the

Office of Technical Services
U. S. Department of Commerce
Washington 25, D. C.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission to the extent that such employee or contractor prepares, handles or distributes, or provides access to, any information pursuant to his employment or contract with the Commission.

UNCLASSIFIED

ORNL-2114

COPY NO.

Contract No. W-7405-eng-26

# STABLE ISOTOPES DIVISION

# SEMIANNUAL PROGRESS REPORT

## For Period Ending May 20, 1956

C. P. Keim, Director
J. R. McNally, Jr., Associate Director

DATE ISSUED

AUG 1 1956

OAK RIDGE NATIONAL LABORATORY
Operated by
UNION CARBIDE NUCLEAR COMPANY
A Division of Union Carbide and Carbon Corporation
Post Office Box P
Oak Ridge, Tennessee

UNCLASSIFIED

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES
3 4456 0350302 8

Reports previously issued in this series are as follows:

| | |
|---|---|
| Y-497 | July 1, 1949 to September 30, 1949 |
| Y-550 | October 1, 1949 to December 31, 1949 |
| Y-586 | January 1, 1950 to March 31, 1950 |
| Y-636 | April 1, 1950 to June 30, 1950 |
| Y-685 | July 1, 1950 to September 30, 1950 |
| Y-723 | October 1, 1950 to December 31, 1950 |
| Y-765 | January 1, 1951 to March 31, 1951 |
| Y-797 | April 1, 1951 to June 30, 1951 |
| Y-821 | July 1, 1951 to September 30, 1951 |
| ORNL-1228 | October 1, 1951 to December 31, 1951 |
| ORNL-1298 | January 1, 1952 to March 31, 1952 |
| ORNL-1356 | April 1, 1952 to June 30, 1952 |
| ORNL-1434 | July 1, 1952 to September 30, 1952 |
| ORNL-1483 | Period Ending December 31, 1952 |
| ORNL-1563 | Period Ending May 20, 1953 |
| ORNL-1617 | Period Ending November 20, 1953 |
| ORNL-1732 | Period Ending May 20, 1954 |
| ORNL-1829 | Period Ending November 20, 1954 |
| ORNL-1908 | Period Ending May 20, 1955 |
| ORNL-2028 | Period Ending November 20, 1955 |

*INTERNAL DISTRIBUTION*

1. C. E. Center
2. Biology Library
3. Health Physics Library
4-6. Central Research Library
7. Reactor Experimental Engineering Library
8-12. Laboratory Records Department
13. Laboratory Records, ORNL R.C.
14. L. B. Emlet (K-25)
15. J. P. Murray (Y-12)
16. A. M. Weinberg
17. E. H. Taylor
18. E. D. Shipley
19. W. H. Jordan
20. M. L. Nelson
21. J. A. Swartout
22. S. C. Lind
23. F. L. Culler
24. A. H. Snell
25. A. Hollaender
26. M. T. Kelley
27. G. H. Clewett
28. K. Z. Morgan
29. T. A. Lincoln
30. A. S. Householder
31. C. S. Harrill
32. C. E. Winters
33. D. W. Cardwell
34. E. M. King
35. D. D. Cowen
36. P. M. Reyling
37. C. P. Keim
38. S. Cromer
39. B. M. Lang (K-25)
40. R. S. Livingston
41. R. F. Hibbs (Y-12)
42. C. E. Normand
43. J. R. McNally, Jr.
44. C. R. Baldock
45. Boyd Weaver
46. G. C. Williams
47. R. W. Johnson
48. E. J. Murphy
49. P. S. Baker
50. M. J. Skinner
51. A. F. Rupp
52. J. L. Gabbard
53. R. A. Charpie
54. G. E. Boyd
55. J. E. Keeton
56. L. O. Love
57. H. R. Gwinn (Y-12)
58. A. E. Cameron
59. A. O. C. Nier (consultant)
60. L. P. Smith (consultant)
61. J. C. Bailar, Jr. (consultant)
62. W. R. Brode (consultant)
63. R. R. Dickison
64. J. S. Drury
65. ORNL – Y-12 Technical Library, Document Reference Section

*EXTERNAL DISTRIBUTION*

66-71. Argonne National Laboratory
72-73. Atomic Energy Commission, Washington
74-76. Brookhaven National Laboratory
77-78. Union Carbide Nuclear Company (K-25)
79. Chief of Naval Research
80. Chicago Patent Group
81-83. duPont Company, Aiken
84. General Electric Company, Richland

85. Hanford Operations Office
86. Iowa State College
87-88. Knolls Atomic Power Laboratory
89. Los Alamos Scientific Laboratory
90. Naval Research Laboratory
91-92. New York Operations Office
93. Patent Branch, Washington
94. Savannah River Operations Office, Augusta
95. duPont Company, Wilmington
96-98. University of California Radiation Laboratory, Berkeley
99-782. Given distribution as shown in TID-4500 (11th ed.) under Chemistry category
783. Division of Research and Development, AEC, ORO

DISTRIBUTION PAGE TO BE REMOVED IF REPORT IS GIVEN PUBLIC DISTRIBUTION

## CONTENTS

PUBLICATIONS ..... vii

MEETINGS AT WHICH PAPERS WERE PRESENTED ..... vii

TRAVELING LECTURES ..... vii

PAPERS CONCERNING USAGE OF ENRICHED ISOTOPES ..... viii

SPECTROSCOPY RESEARCH LABORATORY ..... 1
- Introduction ..... 1
- Molecular Spectroscopy ..... 1
  - Infrared and Microwave Spectra of Formyl and Deutero-Formyl Fluoride ..... 1
  - Infrared Spectrum of TCN ..... 1
  - Molecular-Force-Field Computations by the Oracle ..... 1
  - Vibrational Spectrum of $Ca(OH)_2$ ..... 2
  - Molecular Spectrum of Polonium, $Po_2$ ..... 2
  - Infrared Spectroscopy of Solids ..... 4
- Atomic Spectroscopy ..... 5
  - Spectrum of Singly Ionized Plutonium, Pu(II) ..... 5
  - Atomic Spectrum of Tellurium(II) ..... 6
  - Oracle Routines for the Reduction of Spectroscopic Data ..... 7
  - Magnetic-Field-Strength Measurements by the Use of Nuclear-Magnetic-Resonance Techniques ..... 7
  - Unusual Doppler Effects in Calutron Type of Ion Sources ..... 7
  - Atomic Spectrum of Uranium(II) ..... 9
- X-Ray Spectroscopy ..... 9
  - X-Ray Fluorescence Studies ..... 9
- Spectrochemistry ..... 9
  - General Spectrochemistry ..... 9
  - Rare Earths ..... 10

MASS SPECTROMETRY ..... 11
- Metastable and Collision-induced Transitions in the Mass Spectra of *n*-Butane and *i*-Butane ..... 11
- Ion Beam Attenuation Due to Charge Transfer in the Mass Spectrometer ..... 13
- Experimental Determination of Ion Beam Width in a Mass Spectrometer ..... 13
- Stable Isotope Analyses ..... 16
- Two-Stage Mass Spectrometer MS-F ..... 17
- Mass Spectrometer Assay ..... 17

STABLE ISOTOPE SEPARATION AND CALUTRON OPERATIONS ..... 20
- Introduction ..... 20
- Calutron Operations ..... 20
  - Barium, Series HJ-XAX ..... 20
  - Magnesium, Series HM-XBX ..... 20
  - Tungsten, Series HN-XAX ..... 21
  - Silver, Series HO-XBX ..... 21
  - Germanium, Series HP-XBX ..... 22
  - Cadmium, Series HQ-XAX ..... 22
  - Zinc, Series HR-XBX ..... 23
  - Europium, Series HS-XAX ..... 23
  - Thallium, Series HT-XAX ..... 24
  - Copper, Series HV-XBX ..... 24

Rubidium, Series HW-XAX ..... 25
Rhenium, Series HX-XBX ..... 25
Chlorine, Series HY-XBX ..... 26
Lead, Series HZ-XAX ..... 26
Preparation of Charge Materials ..... 26
Development and Process Improvement ..... 27
Thermal Diffusion ..... 27
Inert Gas Collection in the Electromagnetic Separator ..... 28

STABLE ISOTOPE CHEMISTRY ..... 30
Introduction ..... 30
Chemical Refinement of Isotopes ..... 30
Magnesium ..... 30
Bromine ..... 30
Strontium ..... 30
Molybdenum ..... 31
Silver ..... 32
Samarium ..... 32
Protactinium Chemistry ..... 33
Rare-Earth Chemistry ..... 34
Raw Material Processing ..... 34
Separations by Liquid-Liquid Extraction ..... 34
Process Development ..... 34
Analytical Development ..... 36

UTILIZATION OF ENRICHED STABLE ISOTOPES ..... 38
Target Preparation and Special Services ..... 38
Target Preparation ..... 38
Special Procedures ..... 38
Cooperative Calutron-Cyclotron Program ..... 39
Isotope Distribution ..... 39

## PUBLICATIONS

G. L. Rogosa and W. F. Peed, "Technetium *K* X-Ray Spectrum," *Phys. Rev.* **100**, 1763 (1955).

G. L. Rogosa and W. F. Peed, "*L* X-Ray Spectra of Np, Pu, and Am," *Phys. Rev.* **101**, 591 (1956).

K. L. Vander Sluis and J. R. McNally, Jr., "Fabry-Perot Interferometer with Finite Apertures," *J. Opt. Soc. Amer.* **46**, 39 (1956).

P. A. Staats, H. W. Morgan, and J. H. Goldstein, "Infrared Spectra of $T_2O$, THO, and TDO," *J. Chem. Phys.* **24**, 916 (1956).

## MEETINGS AT WHICH PAPERS WERE PRESENTED

*American Chemical Society, Dallas, Texas, April 9–13, 1956*

Russell Baldock, "The Mass Spectrometer in Inorganic Analysis."

*Southeastern Section of the American Physical Society, Nashville, Tennessee, March 29–31, 1956*

H. M. Rosenstock, "Mass Spectra of Polyatomic Molecules."
P. A. Staats, H. W. Morgan, and J. H. Goldstein, "Infrared Spectra of $T_2O$, THO, and TDO."

*Spectroscopy Symposium, Argonne National Laboratory, February 15–17, 1956*

J. R. McNally, Jr., and M. R. Skidmore, "Unusual Doppler Effects in Calutron Ion Sources."
G. W. Charles, "Vibrational Analysis of the Spectrum of $Po_2$."
P. M. Griffin and K. L. Vander Sluis, "Application of New Techniques to Zeeman Spectroscopy."

*Atomic Energy Commission Information Meeting on Fundamental High-Temperature Chemistry, Oak Ridge National Laboratory, Oak Ridge, Tennessee, May 9–11, 1956*

H. M. Rosenstock, L. K. Brice, J. R. Walton, and Russell Baldock, "Mass Spectra of Silver Halide Vapors."
H. W. Morgan, "Molecular Spectroscopy of the Solid State."

*Fourth Annual Southeastern Seminar on Spectroscopy, University of Florida, Gainesville, January 4–6, 1956*

J. R. McNally, Jr., "High Resolution Spectroscopy."

*Meeting of the Mississippi Academy of Science, Jackson, April 27–28, 1956*

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

*Tennessee Academy of Science, Cookeville, December 2–3, 1955*

P. S. Baker and C. P. Keim, "Radioisotopes from Electromagnetically Enriched Isotopes."

## TRAVELING LECTURES

Russell Baldock, *Current Research in Mass Spectrometry*, Alabama Polytechnic Institute, Auburn, December 16, 1956.

H. M. Rosenstock, *The Mass Spectrometer and High Temperature Chemistry*, Virginia Polytechnic Institute, Blacksburg, March 5, 1956.

H. W. Morgan, *Chemical Applications of Microwave Spectroscopy*, Agricultural and Mechanical College of Texas, College Station, March 21, 1956, and Rice Institute, Houston, Texas, March 23, 1956.

P. M. Griffin, *New Techniques in the Study of Zeeman Effects in Atomic Spectroscopy*, Clemson Agricultural College, Clemson, South Carolina, May 3, 1956, and Virginia Polytechnic Institute, Blacksburg, May 9, 1956.

## PAPERS CONCERNING USAGE OF ENRICHED ISOTOPES

| PUBLICATIONS | ISOTOPES USED |
|---|---|
| L. Grodzins and H. Motz (Brookhaven National Laboratory), "Decay of 72-Second and 49-Day In-114," *Phys. Rev.* **100**, 1236 (1955). | Cd-114 |
| J. M. LeBlanc, L. M. Bollinger, and R. E. Coté (Argonne National Laboratory), "Neutron Transmission Measurements on Br and Se," *Phys. Rev.* **100**, 1248 (1955). | Se-77, -78, -80 |
| W. John (University of California), "Excitation Functions for Alpha-Induced Reactions on Lead," *Phys. Rev.* **100**, 1265 (1955). | Pb-206; Ni-60; Cu-63 |
| E. A. Wolicki, L. W. Fagg, and E. H. Geer (Naval Research Laboratory), "Coulomb Excitation of Energy Levels in Br and Re," *Phys. Rev.* **100**, 1265 (1955). | Br-79, -81; Re-185, -187 |
| B. E. Simmons, K. F. Famularo, and G. D. Freier (University of Minnesota), "Coulomb Excitation of Samarium," *Phys. Rev.* **100**, 1265–66 (1955). | Sm-148, -150, -153, -154 |
| R. H. Davis, A. S. Divatia, R. D. Moffat, and D. A. Lind (University of Wisconsin), "Coulomb Excitation of Re, Ir, and Hg by Protons," *Phys. Rev.* **100**, 1266 (1955). | Re-185, -187; Ir-191, -193; Hg-199, -200 |
| A. S. Divatia, R. H. Davis, R. D. Moffat, and D. A. Lind (University of Wisconsin), "Coulomb Excitation of In-115, I-127, Ta-181, Th-232, and U-238," *Phys. Rev.* **100**, 1266 (1955). | In-115; Ta-181 |
| P. F. Yergin and B. P. Fabricand (University of Pennsylvania), "Sr-86, -87, -88, Y-89, Zr-92 ($\gamma,n$) Cross Sections," *Phys. Rev.* **100**, 1269 (1955). | Sr-86, -87, -88; Zr-92 |
| W. S. Lyon and T. H. Handley (Oak Ridge National Laboratory), "Radioactive Ca-47," *Phys. Rev.* **100**, 1280 (1955). | Ca-46 |
| L. W. Fagg, E. A. Wolicki, R. O. Bondelid, K. L. Dunning, and S. Snyder (Naval Research Laboratory), "Coulomb Excitation of Tellurium and Silver," *Phys. Rev.* **100**, 1299 (1955). | Te-123, -125; Ag-107, -109 |
| J. D. Knight and E. W. Baker (Brookhaven National Laboratory), "Radiochemical Study of Tl-195, Tl-197, and Tl-198," *Phys. Rev.* **100**, 1334–35 (1955). | Hg-196, -198 |
| K. Murakawa (Institute of Science and Technology, Tokyo, Japan), "Nuclear Moments of Mo-95, Mo-97, Zr-91, I-127, Sb-121, and Sb-123," *Phys. Rev.* **100**, 1369–72 (1955). | Zr-91; Sb-121, -123; Mo-95, -97 |
| W. A. Cassatt, Jr., and W. W. Meinke (University of Michigan), "Decay Scheme of In-107," *Phys. Rev.* **100**, 1372–73 (1955). | Cd-106 |
| F. McClung and J. R. Holmes (University of Southern California), "Isotope Shift in the Spectra of Hg-198–Hg-202," *Phys. Rev.* **100**, 1792 (1955). | Hg-198, -202 |
| S. W. Mead, M. D. Petroff, and W. O. Doggett (University of California, Berkeley), "Half-Life of Sn-110," *Phys. Rev.* **100**, 1794 (1955). | Cd-108 |
| J. F. Streib (Stanford University), "Elastic Scattering of Electrons by Li-6 and Li-7," *Phys. Rev.* **100**, 1797 (1955). | Li-6 |
| D. W. Martin, M. K. Brice, J. M. Cork, and S. B. Burson (Argonne National Laboratory), "Decay of Radioactive Ce-143," *Phys. Rev.* **101**, 182 (1956). | Ce-142 |
| J. B. Marion and R. A. Chapman (The Rice Institute), "Low-Lying Level Structure of the Zn-65 Nucleus," *Phys. Rev.* **101**, 282 (1956). | Cu-65 |

| PUBLICATIONS | ISOTOPES USED |
|---|---|
| A. R. Quinton and W. T. Doyle (Sloane Physics Laboratory, Yale University), "($\alpha$,$n$) Type Reactions in Light Nuclei," *Phys. Rev.* **101**, 669 (1956). | B-10 |
| G. F. Pieper and G. S. Stanford (Sloane Physics Laboratory, Yale University), "B-10 + $\alpha$ Reactions," *Phys. Rev.* **101**, 672 (1956). | B-10 |
| W. E. Graves and A. C. G. Mitchell (Indiana University), "Disintegration of Te-129," *Phys. Rev.* **101**, 701 (1956). | Te-128 |
| V. R. Potnis, C. E. Mandeville, and J. S. Burlew (Bartol Research Foundation of The Franklin Institute), "Radiations from Four Radioactive Isotopes of Platinum," *Phys. Rev.* **101**, 753 (1956). | Pt-190, -192, -194, -196 |
| H. B. Willard, J. K. Bair, J. D. Kington, and H. O. Cohn (Oak Ridge National Laboratory), "Elastic Scattering of Neutrons by Li-6 and Li-7," *Phys. Rev.* **101**, 765 (1956). | Li-6, -7 |
| G. Gardner and J. I. Hopkins (Vanderbilt University), "Evidence for the 258-kev Gamma Ray of Sn-113," *Phys. Rev.* **101**, 999–1000 (1956). | Sn-112 |
| M. Awschalom (University of Rochester), "Search for Double Beta Decay in Ca-48 and Zr-96," *Phys. Rev.* **101**, 1041 (1956). | Ca-48; Zr-96 |
| G. D. O'Kelley, N. H. Lazar, and E. Eichler (Oak Ridge National Laboratory), "Decay Chain Ca-49, Sc-49, Ti-49," *Phys. Rev.* **101**, 1059 (1956). | Ca-48 |
| W. E. Graves and S. K. Suri (Indiana University), "Disintegration of Sc-47," *Phys. Rev.* **101**, 1368 (1956). | Ca-44 |
| A. B. Robbins (Sloane Physics Laboratory, Yale University), "Li-7($\alpha$,$n$)B-10 Reaction," *Phys. Rev.* **101**, 1373 (1956). | Li-6, -7 |
| D. R. Speck (University of California), "Hyperfine Structure and Nuclear Moments of Gadolinium," *Phys. Rev.* **101**, 1725 (1956). | Gd-152, -154, -155, -157 |
| D. R. Speck and F. A. Jenkins (University of California, Berkeley), "Nuclear Moments of Hf-177 and Hf-179," *Phys. Rev.* **101**, 1831 (1956). | Hf-177, -179 |
| R. R. Palmer and L. M. Bollinger (Argonne National Laboratory), "Neutron Cross-Section Measurements of Antimony, Gallium, Cadmium, and Mercury," *Phys. Rev.* **102**, 228–30 (1956). | Ga-69, -71; Cd-108, -110, -111, -112, -113, -114, -116; Hg-198, -199, -201; Sb-121, -123 |
| B. L. Cohen (Oak Ridge National Laboratory), "Proton-Induced Reactions in Na-23, Al-27, and Si-29," *Phys. Rev.* **102**, 453–54 (1956). | Si-29 |
| R. M. Sinclair (Westinghouse Research Laboratories), "Gamma Radiation from Inelastic Scattering of Fast Neutrons by Isotopes of Tellurium, Nickel, and Copper," *Phys. Rev.* **102**, 461–64 (1956). | Te-120, -122, -123, -124, -125, -126, -128, -130; Ni-58; Cu-63, -65 |
| B. Saraf (Bartol Research Foundation of The Franklin Institute), "Inner Bremsstrahlung Emitted in Electron Capture Decay of Ni-59, A-37, and Fe-55," *Phys. Rev.* **102**, 466–67 (1956). | Ni-58; Fe-54 |
| L. Grodzins and H. Motz (Brookhaven National Laboratory), "Decay of In-114$m$ and In-114," *Phys. Rev.* **102**, 761 (1956). | Cd-114 |
| R. K. Golden and S. Frankel (University of Pennsylvania), "Electron Scattering in the $K$-Electron – Gamma Angular Correlation in Sn-117," *Phys. Rev.* **102**, 1053 (1956). | Sn-116 |
| C. E. Weller and J. C. Grosskreutz (University of Texas), "Gamma Rays from the Proton Bombardment of Separated Copper Isotopes," *Phys. Rev.* **102**, 1149 (1956). | Cu-63, -65 |

| PUBLICATIONS | ISOTOPES USED |
|---|---|
| K. B. Adams and K. Burns (Westinghouse Research Laboratories – Allegheny Observatory), "Vacuum Interferometer and Cadmium Oven," *J. Opt. Soc. Amer.* **46**, 36 (1956). | Cd-114 |
| L. T. Aldrich (Carnegie Institution of Washington), "Measurement of Radioactive Ages of Rocks," *Science* **123**, No. 3203, 871–75 (1956). | K-40; Rb-87 |
| J. P. Schiffer, C. R. Gossett, G. C. Phillips, and T. E. Young (The Rice Institute), "Energy Levels in Cl-35 and Cl-37," *Bull. Am. Phys. Soc.* (Ser. II) **1**(2), 95 (1956). | Cl-35, -37 |
| R. A. Chapman, J. B. Marion, and J. C. Slattery (The Rice Institute), "Thresholds in the Reactions Mn-55($p,n$)Fe-55 and Zn($p,n$)Ga," *Bull. Am. Phys. Soc.* (Ser. II) **1**(2), 95 (1956). | Zn-67, -68 |
| H. J. Watters (Massachusetts Institute of Technology), "Elastic and Inelastic Scattering of 31-Mev Alpha Particles from Light Nuclei," *Bull. Am. Phys. Soc.* (Ser. II) **1**(1), 20 (1956). | -6; C-12; Mg-24 |
| G. W. Wetherill, L. T. Aldrich, G. R. Tilton, and G. L. Davis (Carnegie Institution of Washington), "Half-life of Rb-87," *Bull. Am. Phys. Soc.* (Ser. II) **1**(1), 31 (1956). | Rb-87 |
| M. Awschalom (University of Rochester), "Lower Limits for the Half-Lives of Ca-48 and Zr-96, Against Double Beta Decay," *Bull. Am. Phys. Soc.* (Ser. II) **1**(1), 31 (1956). | Ca-48; Zr-96 |
| M. S. Moore, J. P. Schiffer, and C. M. Class (The Rice Institute), "Excitation Curve and Angular Distributions for the Ni-58($p,p'\gamma$1.45)Ni-58 Reaction," *Bull. Am. Phys. Soc.* (Ser. II) **1**(1), 39–40 (1956). | Ni-58 |
| C. R. Gossett, J. W. Butler, and H. D. Holmgren (Naval Research Laboratory), "Proton Capture Resonances in Ni-58 and Ni-60," *Bull. Am. Phys. Soc.* (Ser. II) **1**(1), 40 (1956). | Ni-58, -60 |
| R. M. Sinclair (Westinghouse Research Laboratories), "Gamma Radiation from the Interaction of 4.4 Mev Neutrons with Isotopes of Tellurium, Copper, and Nickel," *Bull. Am. Phys. Soc.* (Ser. II) **1**(1), 42 (1956). | Te-125, -126, -128, -130; Ni-58; Cu-63, -65 |
| J. R. Huizenga, V. E. Krohn, and S. Raboy (Argonne National Laboratory), "Angular Correlation of Gamma Rays in Pb-204$m$," *Bull. Am. Phys. Soc.* (Ser. II) **1**(1), 43 (1956). | Pb-204 |
| A. Taylor, H. Marshak, and H. W. Newson (Duke University), "Total Cross Sections in the Kilovolt Region; Mg-24, Mg-25, Mg-26," *Bull. Am. Phys. Soc.* (Ser. II) **1**(1), 62 (1956). | Mg-24, -25, -26 |
| A. W. Schardt and B. J. Dropesky (Los Alamos Scientific Laboratory), "Decay of V-53 and V-54," *Bull. Am. Phys. Soc.* (Ser. II) **1**(4), 162 (1956). | Cr-53, -54 |
| J. D. Kurbatov, H. J. Sathoff, Jr., K. Hisatake, and M. Sakai (The Ohio State University), "Beta Spectrum of Co-56," *Bull. Am. Phys. Soc.* (Ser. II) **1**(4), 163 (1956). | Fe-56 |
| J. W. Butler, C. R. Gossett, and H. D. Holmgren (Naval Research Laboratory), "Beta Decay of Cu-59," *Bull. Am. Phys. Soc.* (Ser. II) **1**(4), 163 (1956). | Ni-58 |
| F. K. McGowan, P. H. Stelson, and M. M. Bretscher (Oak Ridge National Laboratory), "Angular Distribution of Gamma Rays Following Coulomb Excitation in Pt-194, Cd-114, Pd-110, Ru-104, Cd-113, and Cd-111," *Bull. Am. Phys. Soc.* (Ser. II) **1**(4), 164 (1956). | Pt-194; Pd-110; Ru-104; Cd-111, -113, -114 |

| PUBLICATIONS | ISOTOPES USED |
|---|---|
| P. H. Stelson and F. K. McGowan (Oak Ridge National Laboratory), "Polarization of Gamma Rays Following Coulomb Excitation," *Bull. Am. Phys. Soc.* (Ser. II) 1(4), 164 (1956). | Ag-107, -109; Cd-111, -113 |
| N. P. Heydenburg and G. M. Temmer (Carnegie Institution of Washington), "Coulomb Excitation of V-50, Kr, Xe, Te, and Eu-151," *Bull. Am. Phys. Soc.* (Ser. II) 1(4), 164–65 (1956). | V-50; Te-120, -122, -124, -126, -128, -130 |
| L. W. Fagg, E. H. Geer, and E. A. Wolicki (Naval Research Laboratory), "Coulomb Excitation of V, Ni, Ga, Rb, and Sn," *Bull. Am. Phys. Soc.* (Ser. II) 1(4), 165 (1956). | V-50; Ni-61; Sn-115, -117, -119 |
| R. M. Brugger, J. E. Evans, E. G. Joki, and R. S. Shankland (Phillips Petroleum Company), "Total Neutron Cross Section of Chlorine, 0.03 to 15,000 ev.," *Bull. Am. Phys. Soc.* (Ser. II) 1(4), 176 (1956). | Cl-35 |
| R. B. Schwartz, V. E. Pilcher, and R. M. Schectman (Brookhaven National Laboratory), "Slow Neutron Resonances in Cd and W," *Bull. Am. Phys. Soc.* (Ser. II) 1(4), 187 (1956). | Cd-113; W-182, -183 |
| G. M. Frye, Jr., and J. H. Gammell (Los Alamos Scientific Laboratory), "The B-10($n,t2\alpha$) and B-10($n,dn2\alpha$) Reactions for 6- to 20-Mev Neutrons," *Bull. Am. Phys. Soc.* (Ser. II) 1(4), 196 (1956). | B-10 |
| E. Der Mateosian and M. Goldhaber (Brookhaven National Laboratory), "Isomeric Transition in Cd-113*m*," *Bull. Am. Phys. Soc.* (Ser. II) 1(4), 207 (1956). | Cd-112 |
| A. Sperduto and W. W. Buechner (Massachusetts Institute of Technology), "($d,p$) Reactions of the Iron Isotopes," *Bull. Am. Phys. Soc.* (Ser. II) 1(4), 223 (1956). | Fe-54, -57, -58 |
| S. S. Markowitz, F. S. Rowland, and G. Friedlander (Princeton University and Brookhaven National Laboratory), "($p,pn$) Excitation Functions from 0.4 to 3.0 Bev.," *Bull. Am. Phys. Soc.* (Ser. II) 1(4), 224 (1956). | Fe-54; Ni-68; Cu-63; Zn-64 |

# STABLE ISOTOPES DIVISION SEMIANNUAL PROGRESS REPORT

## SPECTROSCOPY RESEARCH LABORATORY

J. R. McNally, Jr.

### INTRODUCTION

The activities of the Spectroscopy Research Laboratory include research and development in the fields of molecular, atomic, and x-ray spectroscopy. This work is directed toward the determination of fundamental spectroscopic data and the investigation of new instrumental methods of analysis for isotopes, elements, and compounds.

Major accomplishments during this semiannual period are as follows:

1. A new improved technique has been developed for the study of the vibrational spectra of "free" molecules in normally solid systems.
2. Tritium cyanide (TCN) has been synthesized and its fundamental infrared vibrations have been observed.
3. An unusual Doppler effect has been observed in the spectra of ions excited in the calutron J arc.
4. A direct analytical technique involving x-ray fluorescence determinations of yttrium has been developed.

### MOLECULAR SPECTROSCOPY

#### Infrared and Microwave Spectra of Formyl and Deutero-Formyl Fluoride

P. A. Staats H. W. Morgan

A. H. Nielsen and R. Stratton, of the University of Tennessee, are continuing the study of the infrared spectra at high resolution, using compounds supplied by this laboratory. The experimental work is approximately 50% complete. Preliminary analyses of the data indicate that the rotational parameters will be evaluated with good accuracy. It is expected that values of these parameters will permit an interpretation of the microwave spectra previously observed.

#### Infrared Spectrum of TCN

P. A. Staats H. W. Morgan
J. H. Goldstein[1]

The infrared spectrum of TCN gas was studied in the region 4000 to 400 $cm^{-1}$ with a Perkin-Elmer single-beam double-pass spectrometer equipped with LiF, NaCl, and KBr prisms. Approximately 0.005 mole (130 curies) of TCN was prepared, giving a maximum pressure of 30 cm Hg in a 10-cm path-length absorption cell.

The TCN was made by the reaction of $T_2O$ vapor with solid $P(CN)_3$; $T_2O$ was synthesized in a manner previously described.[2] The $P(CN)_3$ was prepared by the reaction[3] of AgCN and $PCl_3$ and was employed primarily because its sublimation at about 170°C affords a simple and effective purification. The $T_2O$ vapor was brought in contact with an excess of $P(CN)_3$ at room temperature, and gentle heat was then applied to ensure complete reaction. Recovery of the tritium was accomplished by the use of metallic uranium, as has been previously described.

In the gas phase, under the above conditions of pressure and volume, TCN displays a spontaneous polymerization. Its rate is such that after 12 hr approximately 80% of the sample has disappeared. When the reaction is allowed to go to completion, only a small pressure of $T_2$ gas remains. It is tentatively believed that free radicals produced by the beta activity initiate polymer formation.

The frequency data are summarized in Table 1. The observed TCN frequencies are considered accurate to within ±2 $cm^{-1}$. Data on HCN and DCN are included for comparison. The anharmonicity corrections appear to be consistent. That for $\nu_3$ is very nearly inversely proportional to the reduced mass computed for the H–CN stretch, treated as a diatomic case. The corrections for $\nu_1$ are reasonably constant, while those for $\nu_2$ depend inversely on $\nu_2^2$. These trends are in accord with the nature of the normal modes in these molecules.

#### Molecular-Force-Field Computations by the Oracle

H. W. Morgan J. H. Goldstein

General consideration of the problem has confirmed the belief that the evaluation of a diagonal *F*-matrix is best achieved by a converging, iterative technique of the type used previously on the

[1]Consultant, Emory University, Atlanta, Georgia.

[2]P. A. Staats, H. W. Morgan, and J. H. Goldstein, *J. Chem. Phys.* 24(4), 916 (1956).

[3]H. Hübner and G. Wehrhane, *Ann. Chem., Justus Liebigs* 128, 254 (1863); 132, 277 (1864).

TABLE 1. VIBRATIONAL SPECTRA OF HYDROGEN CYANIDES

| Molecule | Assignment | Observed Frequency | Anharmonicity Correction | $\omega_{obs}$ [a] | $\omega_{calc}$ [b] |
|---|---|---|---|---|---|
| HCN[c] | $\nu_1$ | 2095.5 (Raman) | 29.2 | 2124.7 | 2125.6 |
| | $\nu_2$ | 711.7 | 15.0 | 726.7 | |
| | $\nu_3$ | 2211.7 | 134.5 | 3446.2 | 3448.8 |
| DCN[c] | $\nu_1$ | 1928 | 25.4 | 1953 | 1955.8 |
| | $\nu_2$ | 568.9 | 10.7 | 579.6 | 579.5 |
| | $\nu_3$ | 2629.3 | 70.94 | 2700.2 | 2700.3 |
| TCN | $\nu_1$ | 1724 | 27.5[d] | | 1751.9 |
| | $\nu_2$ | 513 | 8.4[d] | | 521.4 |
| | $\nu_3$ | 2460 | 47.2[d] | | 2507.2 |

[a]Fundamental frequency ($cm^{-1}$).
[b]Calculated by using $\delta_{ij}$ of ref. 3.
[c]Reported by A. E. Douglas and D. Sharma, *J. Chem. Phys.* 21, 448 (1953).
[d]Obtained from $\omega_{calc}$ minus observed frequency.

formyl fluoride problem. Several schemes have been proposed for the use of this technique in determining off-diagonal elements. The writing of the complete Oracle code for this problem is now awaiting revision of the necessary matrix subroutines by members of the Mathematics Panel.

### Vibrational Spectrum of $Ca(OH)_2$

W. R. Busing[4] H. W. Morgan

It has been reported[5] that the infrared spectrum of brucite, crystalline $Mg(OH)_2$, consists of a complex pattern of frequencies associated with the OH stretching vibration. No satisfactory explanation of this spectrum has been given. Recently, the availability of single crystals of $Ca(OH)_2$, together with precise structural data on the unit cell obtained by neutron diffraction,[6] led to the examination of the infrared vibrational spectrum of this compound in the hope that this fine structure might be interpreted.

The infrared spectra of single crystals of $Ca(OH)_2$ were observed, and fine-structure centering about the characteristic OH stretching frequency was resolved. A number of single crystals were employed, ranging down to 10 $\mu$ in thickness. The fine structure was found to cover a region of approximately 1000 $cm^{-1}$. It was studied at 25°C and at −196°C and with the O−H valence-bond axis oriented from 0 to 45 deg with respect to the incident radiation. The observed spectra are summarized in Figs. 1 and 2.

Based on the frequency and intensity data, an energy-level scheme consistent with the appropriate selection rules was devised to explain the spectrum. Work is in progress to obtain the Raman spectra of single crystals at various orientations in order to investigate the low frequency regions and complete the experimental data.

### Molecular Spectrum of Polonium, $Po_2$

G. W. Charles

The consideration of the principal band system of diatomic polonium was completed, and a manuscript has been prepared for publication. A least-squares fit of the empirical energy levels yielded the constants of the system; the values are given in Table 2. Altogether, 352 bands have been classified in this system in $Po^{210}$-$Po^{210}$ and 316 bands in $Po^{208}$-$Po^{208}$. The dissociation energy of the lower state was calculated to be 1.895 v on the assumption that the dissociation products are two polonium atoms in their normal states $^3P_2'$.

[4]Chemistry Division.

[5]R. T. Mara and G. B. B. M. Sutherland, *J. Opt. Soc. Amer.* 43, 1100 (1953).

[6]W. R. Busing and H. A. Levy, *Chem. Semiann. Prog. Rep. June 30, 1955*, ORNL-1940, p 43.

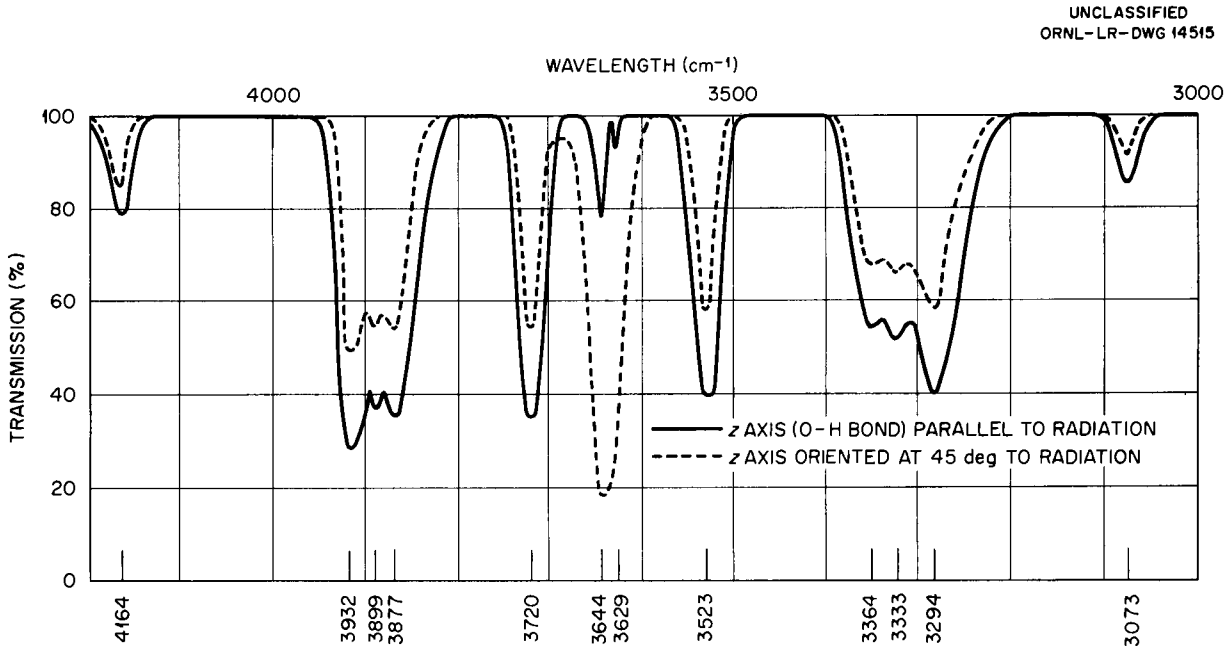



**Fig. 1. Infrared Spectrum of $Ca(OH)_2$ Single Crystal with the $z$ Axis Oriented from 0 to 45 deg.**

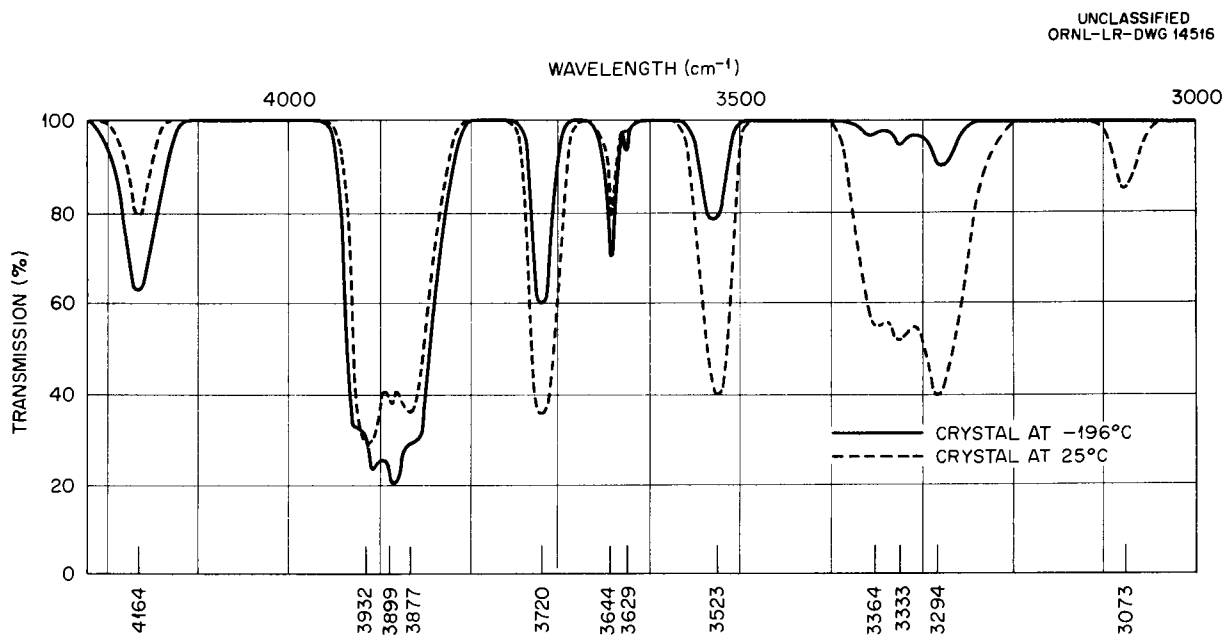



**Fig. 2. Infrared Spectrum of $Ca(OH)_2$ Single Crystal at 25°C and −196°C.**

TABLE 2. SUMMARY OF CONSTANTS OF PRINCIPAL SYSTEM OF POLONIUM

| Molecule | State | $T_e$ (cm$^{-1}$)[a] | $\omega_e$ (cm$^{-1}$)[b] | Probable Error (cm$^{-1}$) | $\omega_e x_e$ (cm$^{-1}$)[c] | Probable Error (cm$^{-1}$) | $\omega_e y_e$ (cm$^{-1}$)[c] | Probable Error (cm$^{-1}$) |
|---|---|---|---|---|---|---|---|---|
| $Po^{210}$-$Po^{210}$ | Lower | 0.0 | $155.71_5$ | 0.03 | $-0.335_3$ | 0.002 | $-0.00032_3$ | 0.00003 |
| $Po^{210}$-$Po^{210}$ | Upper | 25,149.3 | $108.53_2$ | 0.04 | $-0.441_7$ | 0.002 | | |
| $Po^{208}$-$Po^{208}$ | Lower | 0.0 | $156.44_0$ | 0.03 | $-0.340_0$ | 0.002 | $-0.00029_5$ | 0.00003 |
| $Po^{208}$-$Po^{208}$[d] | Lower | | $156.46_2$ | | $-0.338_5$ | | $-0.00032_7$ | |
| $Po^{208}$-$Po^{208}$ | Upper | 25,149.1 | $108.97_7$ | 0.05 | $-0.442_3$ | 0.002 | | |
| $Po^{208}$-$Po^{208}$[d] | Upper | | $109.05_3$ | | $-0.445_9$ | | | |

[a]Electronic energy.
[b]Vibrational frequency.
[c]Anharmonicity constants.
[d]Calculated from the observed constants of $Po^{210}$-$Po^{210}$.

Consideration of the remaining unclassified bands is continuing. Full details on the spectra will be published.[7]

### Infrared Spectroscopy of Solids

H. W. Morgan

The methods of matrix computation being developed give promise of allowing the evaluation of off-diagonal *F*-matrix elements from the shifts observed in isotopic vibrational spectra. Except for elements of low atomic weight, however, such shifts are small and difficult to measure. The problem lies not so much in the resolution of commercial prism spectrographs ($\sim$1 cm$^{-1}$) as in the inherent breadth of the spectrum lines. Thus, consideration has been given to means whereby the molecular environment might be controlled and the lines sharpened.

From a study of the molecular interactions and fine structure responsible for line width, it was concluded that only in the solid state could the proper environments be achieved. In the solid state, line broadening results from the coupling of a given vibration with lattice frequencies and with identical frequencies in neighboring molecules or ions. By the use of solid solutions, with careful choice of solvent, it should be possible to minimize these interactions and to achieve relatively sharp absorptions.

In the experimental work to date the alkali halides have been employed as solvents because of their strongly ionic lattices and because of the advantages of the pressed-window technique. The sample was added to alkali halide powder, and the mixture fused. On cooling, the mixture was ground and pressed under vacuum into a transparent window for study. The relatively high melting points of these solvents (600 to 800°C) appear favorable for the formation of solid solutions.

In the above manner, $Li_3N$ (Fig. 3), $SeO_2$, and a variety of inorganic compounds containing the $CN^-$, $SO_4^{--}$, and $UO_2^{++}$ ions were studied. In each case an increase in absorption intensity and a narrowing of the line width were noted, the spectrum of the pure solid sample being used for comparison. As was expected, these characteristics appear to be determined by the solvent employed. In the case of $SeO_2$, a chain polymer in its solid state, the solid solutions obtained at high temperatures show the spectrum of the monomeric species.

[7]G. W. Charles *et al.*, *Description and Vibrational Analysis of the Molecular Spectrum of Polonium*, ORNL-2118 (to be published).

UNCLASSIFIED
ORNL—LR—DWG 14517

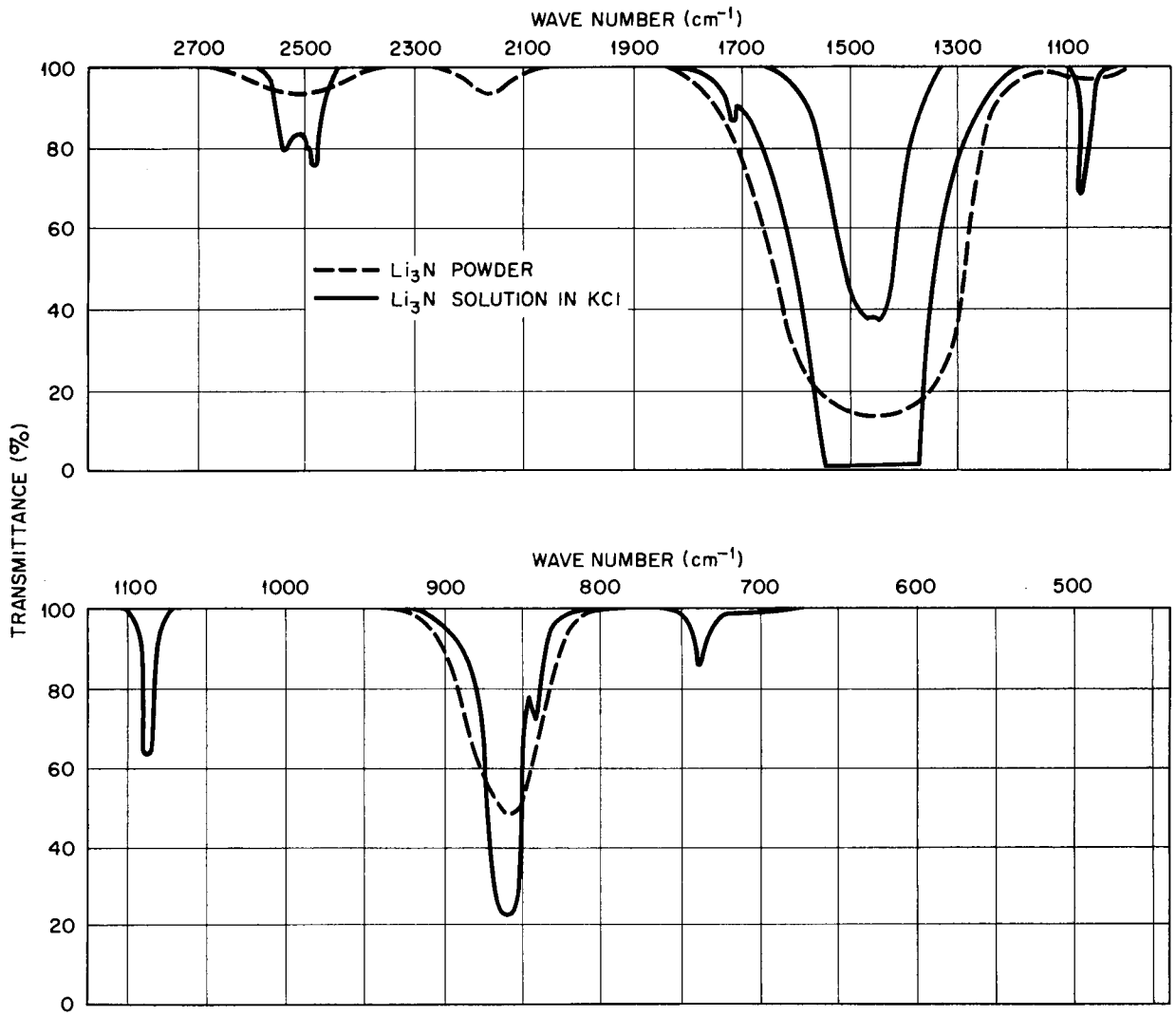



**Fig. 3. Infrared Spectrum for $Li_3N$.**

Some information about the nature of the solid solution was gained by observing crystal growth in the pressed windows. After their preparation, the windows retained an internal pressure, and the KBr solvent slowly recrystallized at room temperature to lessen the strain. The rate of recrystallization can be accelerated by heating; the crystals can be observed with a microscope. The result is a change in the spectrum as the solute molecules or ions are swept out of the solvent lattice. In pressed KBr windows this phenomenon appears to be responsible for the well-known decrease in transmission with time.

Future studies are designed to determine the applicability of this solid solution technique to analytical problems and to study the possibilities of the method in the measurement of small isotope shifts.

## ATOMIC SPECTROSCOPY

### Spectrum of Singly Ionized Plutonium, Pu(II)

J. R. McNally, Jr. P. M. Griffin

Data on the spectra of plutonium ($z = 94$) have been reported in the literature by a number of

investigators. The first is probably that of G. K. Rollefson and H. W. Dodgen, who reported[8] (in July of 1943) 161 of 176 lines as being attributed to the plutonium in a 20-μg sample. The sample was dissolved in 4 *N* HCl, deposited on silver electrodes, and excited with a condensed spark. Subsequently, Dodgen, Chrisney, and Rollefson[9] reported a total of 924 lines in the range 2100 to 7240 Å on an intensity scale of 0 to 10 with an estimated limit of error of ±0.2 Å. In this work they used a 500-μg sample (originating from Oak Ridge) that was excited on silver electrodes in a helium atmosphere by a condensed spark and photographed with a 3-meter Paschen spectrograph having a dispersion of 5.5 Å/mm.

Later, Tomkins and Fred,[10] using 100-μg, or smaller, samples, reported on 229 lines of plutonium as observed by the copper-spark method. Subsequently, data on the following were published: the furnace spectrum, by Conway;[11] the hollow cathode spectrum (714 lines) and hyperfine structure of $Pu^{239}$, by Van den Berg and Klinkenberg;[12] the isotope shifts (238, 239, 240, 242), by Conway and Fred;[13] the hyperfine structure, by Kaliteevskii and Chaika;[14] the hyperfine structure, by Korostyleva *et al.*;[15] the isotope shift (239 and 240), by Striganov *et al.*;[16] and the Zeeman effect, by Griffin and McNally.[17]

Striganov *et al.*[16] conjectured on the assignment of electronic configurations based on hyperfine and isotope shift data, while Van den Berg and Klinkenberg[12] attempted to ascribe ionization stages to the spectrum lines based on excitation data. In both instances rather significant errors exist, as proved by the ORNL Zeeman effects and preliminary classification.[17] The importance of Zeeman effects to the complete interpretation of the spectra of plutonium has been recognized by both Russian and Netherland investigators.

Development of a precision coordinate comparator for the measurement of echellograms[18] and of mathematical routines for least-squares treatment of the measurement data[19] by the Oracle has permitted the remeasurement and re-evaluation of the ORNL Zeeman data with a significant improvement in precision. More than 180 Zeeman patterns, involving *J* values as high as 11/2, have been measured. It is anticipated that over 400 Zeeman patterns on existing spectrograms taken at 24,000 oersteds will be used in extending the spectrum classification.

It has become evident already that, except possibly for unpublished wavelength data in a limited spectrum region obtained by Stukenbroeker[20] at ORNL, the published wavelengths are quite discordant and that a complete precision wavelength study of this spectrum is very much needed. Wavelength differences as large as 0.35 Å and an average bias of 0.11 Å exist between the published data of the Netherlands and Russian groups (see Table 3).

### Atomic Spectrum of Tellurium(II)

P. M. Griffin K. L. Vander Sluis

Work is continuing on the measurement and reduction of the Zeeman spectrum of tellurium. To date, 233 patterns have been measured; 138 of the patterns have been reduced, by the least-squares method,[19] by the Oracle. No extensive work on the analysis of the energy-level systems in Te(II) will be carried out until all the recorded

[8] G. K. Rollefson and H. W. Dodgen, *Report on Spectrographic Analysis Work*, CK-812 (July 1943) (declassified Dec. 29, 1954).

[9] H. W. Dodgen, J. Chrisney, and G. K. Rollefson, *The Spectrum of Plutonium*, CK-2247 (Oct. 9, 1944) (declassified April 20, 1954).

[10] F. S. Tomkins and M. Fred, *Spectra of the Heavy Elements*, ANL-4018 (Aug. 8, 1947) (declassified Feb. 1, 1956); see also *J. Opt. Soc. Amer.* **39**, 357 (1949).

[11] J. G. Conway, *J. Opt. Soc. Amer.* **44**, 276 (1954).

[12] M. Van den Berg and P. F. A. Klinkenberg, *Physica* **20**, 461 (1954); see also p 37.

[13] J. G. Conway and M. Fred, *J. Opt. Soc. Amer.* **43**, 216 (1953).

[14] N. I. Kaliteevskii and M. P. Chaika, *Doklady Akad. Nauk S.S.S.R.* **103**, 49 (1955).

[15] L. A. Korostyleva, A. R. Striganov, and N. M. Iashin, *Zhur. Eksptl. i Teort. Fiz.* **28**, 471 (1955).

[16] A. R. Striganov, L. A. Korostyleva, and Iu. P. Dontsov, *Zhur. Eksptl. i Teort. Fiz.* **28**, 480 (1955).

[17] P. M. Griffin and J. R. McNally, Jr., Proceedings of Rydberg Centennial Conference on Atomic Spectroscopy, Lund University, Lund, Sweden (1955); *J. Opt. Soc. Amer.* **45**, 63 (1955).

[18] P. M. Griffin, *Stable Isotope Research and Production Semiann. Prog. Rep. Nov. 20, 1954*, ORNL-1829, p 9.

[19] K. L. Vander Sluis, P. M. Griffin, and J. H. Vander Sluis, *Stable Isotopes Research and Production Semiann. Prog. Rep. Nov. 20, 1955*, ORNL-2028, p 6.

[20] G. L. Stukenbroeker, G. K. Werner, and B. L. Pope, *Quar. Prog. Rep. Stable Isotope Research and Production Division July 1, 1951 to September 30, 1951*, Y-821, p 31.

**TABLE 3. COMPARISON OF RECENT WAVELENGTH DATA ON PLUTONIUM**

| | Laboratory | |
|---|---|---|
| | Netherlands*–Russian* | Netherlands*–ORNL** |
| Net bias, Å | +0.11 | −0.013 |
| Maximum discrepancy, Å | +0.35 | −0.11 |
| Average deviation, Å | ±0.13 | ±0.023 |

*Measurements made with a 3-prism glass Steinhell spectrograph.

**Measurements made with a 21-ft, 30,000 lines-per-inch Paschen grating spectrograph.

patterns have been measured and the Landé $g$ factors with their probable errors have been calculated.

### Oracle Routines for the Reduction of Spectroscopic Data

P. M. Griffin K. L. Vander Sluis
J. H. Vander Sluis[21]

The Oracle routine for the calculation of laboratory wavelength and vacuum wave numbers[19] from the order number and coordinate comparator readings on a spectrum line as photographed with an Echelle-Littrow spectrograph have been debugged and put in the Oracle routines library. The routine for the calculation of Landé $g$ factors[18] with their probable errors from the raw data from an echellogram has also been debugged and placed in the Oracle library.

### Magnetic-Field-Strength Measurements by the Use of Nuclear-Magnetic-Resonance Techniques

O. B. Rudolph

The determination of the magnetic field strength in the Zeeman magnet is of extensive importance in Zeeman spectroscopy. Although the field strength may be calculated by the measurement of the Zeeman splittings of lines of impurity elements involving levels with known $g$ factors, some Zeeman studies at high precision require an independent measurement of the field.

[21]Mathematics Panel.

Extensive work has been done on the development of a nuclear-magnetic-resonance method of field calibration. Apparatus is commercially available for nuclear-magnetic-resonance field measurements but is not applicable in this problem because of the large size of its probe. The gap in the Zeeman magnet is only ¼ in. by a 1-in. diameter; the region of interest is a "pill box" 3 mm in diameter and 0.1 mm in height, with its axis perpendicular to the field. The small volume to be investigated and the dimensional limitations of the gap introduce many serious experimental difficulties.

With the use of an experimental setup such as that shown in Fig. 4 and an oscillator-detector unit as described by Watkins,[22] good results have been obtained up to 15,000 oersteds and moderate results up to 27,000 oersteds; no signal has been obtained at fields greater than 27,000 oersteds. Measurements at field strengths up to 33,500 oersteds that are accurate to 1 part in 10,000 are needed.

### Unusual Doppler Effects in Calutron Type of Ion Sources

J. R. McNally, Jr. M. R. Skidmore

Spectroscopic studies were made of a low voltage, low pressure, magnetically constrained axial arc (Fig. 5) similar to that used in the ORNL isotope separators (calutrons) as an ion source. Spectrum lines of ionized helium and carbon and recombination lines of hydrogen show unusually marked broadening and a "slant-line effect." The latter effect (Fig. 6) is attributed to a Doppler shift, which results from ordered ionic rotations in the arc column, the ion velocities at times exceeding $10^6$ cm/sec.

Ions moving with a velocity $\vec{v}$ in a magnetic field experience the Lorentz force and describe a spiral motion, the radius of which is

$$\vec{R} = \frac{c}{qH^2} m\vec{v} \times \vec{H} ,$$

where $q$ is the degree of ionization. Additional forces acting on the ion produce a momentum

[22]G. D. Watkins, Thesis, Harvard University, *An R. F. Spectrometer with Application to Studies of Nuclear Magnetic Resonance Absorption in Solids* (1952).

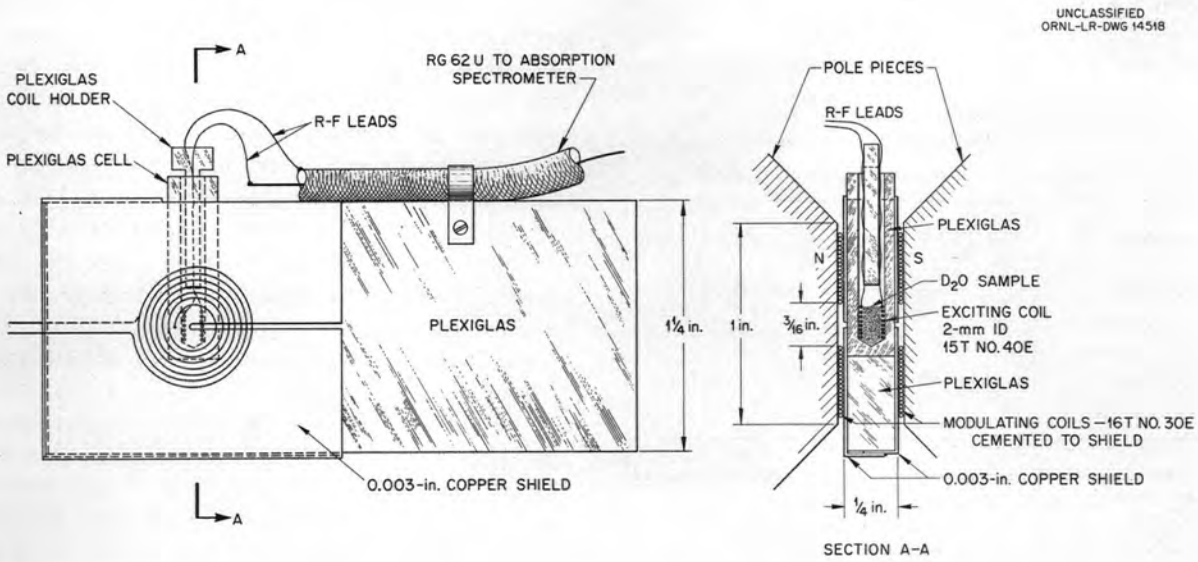



**Fig. 4. Nuclear-Resonance Probe for a ¼-in. Zeeman Magnet Gap.**

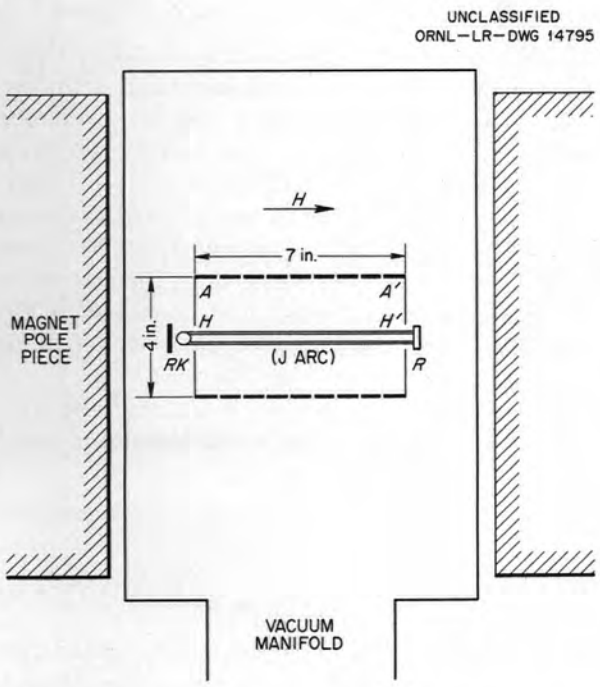



**Fig. 5. Schematic Diagram of J Arc.** *R*, reflection; *K*, filament; *H*,*H′*, collimating holes; *A*,*A′*, anode plates; magnetic fields, up to 7000 oersteds.

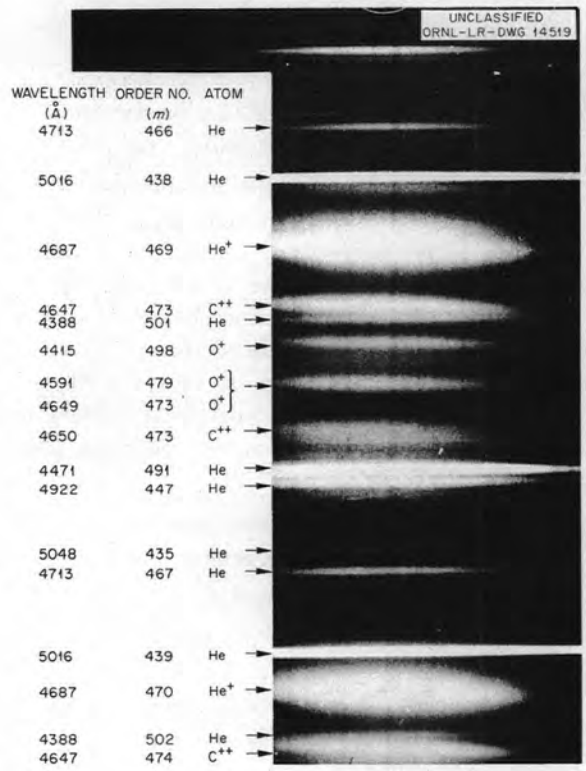



**Fig. 6. Echellogram of J Arc.** Magnetic field, 6600 oersteds; *K* to *A* current, 19 amp. *K* to *A* voltage, 100 v. Note Doppler slant and broadening effects of lines due to ionized atoms.

change and hence a change in $\vec{R}$:

$$\frac{d\vec{R}}{dt} = \frac{c}{qH^2}\,\vec{F} \times \vec{H} = \vec{v}_D \ ,$$

where

$$\vec{F} = \frac{\partial v}{\partial t} + \vec{v}\cdot\nabla\vec{v} \ .$$

Thus, there is produced a drift velocity normal to $H$ and $F$. For the arc under consideration there are at least two obvious forces which may contribute to the observed motion: electrical gradients, which require specific charge distributions, and pressure gradients, which require collision phenomena and ionic density gradients. Thus

$$\vec{F} = q\vec{E} - \frac{\vec{\text{grad}}\ p}{\mu} + \ldots \ .$$

The dependence of $\vec{F}$ and hence $\vec{v}_D$ on the degree of ionization should permit a unique selection of the causal agent if these are the only two forces of significance. A study of spectrum lines of atoms in various stages of ionization [e.g., C(II), C(III), etc.] should permit this evaluation.

### Atomic Spectrum of Uranium(II)

G. W. Charles

An investigation of the spectra of uranium has been initiated. Work is now proceeding on the first phase, a verification of the existing analysis. An extension and improvement of the existing Zeeman data is planned as a part of this phase of the investigation. In preparation, a list of allowed lines of U(II) has been calculated from published energy levels[23] and has been checked against the line list of the uranium spectra compiled by McNally.[24] Zeeman data obtained by McNally while at MIT are being scanned for additional useful information.

[23] J. J. Katz and E. Rabinowitch, *The Chemistry of Uranium*, chap. 2, McGraw-Hill, New York, 1951.

[24] *Wavelength Table of Uranium Spectrum Lines in the Range 11,908 Å to 2025 Å*, compiled by J. R. McNally, Jr. (1950) from data of G. R. Harrison, Massachusetts Institute of Technology, and C. C. Kiess, National Bureau of Standards (material, which has not been published, in files of J. R. McNally).

## X-RAY SPECTROSCOPY

### X-Ray Fluorescence Studies

W. F. Peed

In support of the rare-earth activities of the Division, x-ray methods for analysis of yttrium have been investigated. The limit of detection for liquid samples is 0.1 g/liter; in solids, 0.1%. In concentrations greater than a few per cent, precision of 2% of the amount present can be realized. The method is based on measurement of the intensity ratio of yttrium $K$ alphas to uranium $L$ alphas, the uranium being added as an internal standard (see Fig. 7).

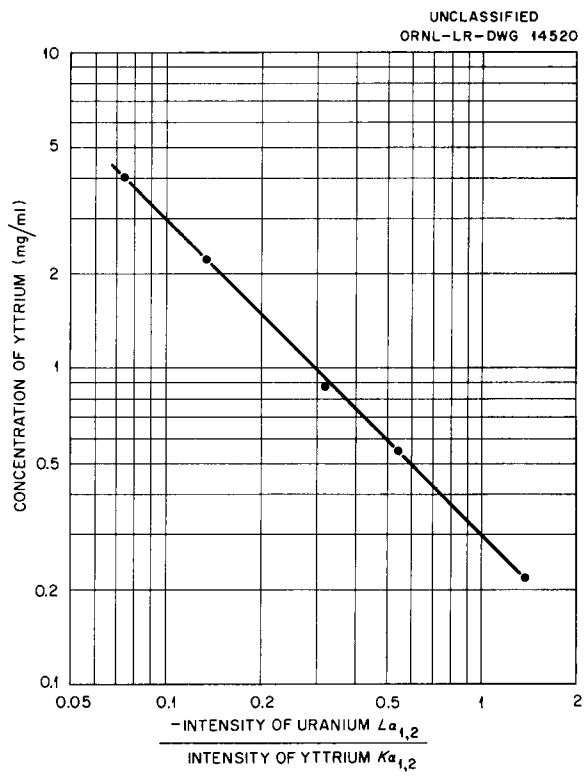



**Fig. 7. X-Ray Fluorescence Data on Yttrium and Uranium in Solutions.**

## SPECTROCHEMISTRY

### General Spectrochemistry

Z. Combs J. J. Mundzak
J. A. Norris

Standards were prepared for the analysis of magnesium oxide at the ppm level for 24 constituent elements.

Further work has been done on the $Y_2O_3$ carrier method for rare earths in metals. The chemical procedure used by the Analytical Chemistry Division shows average recoveries of from 80 to 120% of rare earths added to base $Y_2O_3$. Additional experience, both chemical and spectrographic, should improve the precision of this method. Several steels were checked by the method, and no rare earths were found.

A total of 1194 samples comprising 15,526 determinations was completed by spectrochemical, x-ray, and infrared methods of analysis.

**Rare Earths**

J. J. Mundzak S. J. Ovenshine J. A. Norris

Considerable difficulty has been experienced recently with porous-cup electrodes, and a new "spark-in-spray" method[25] is being examined. This method should also allow direct use of various organic solutions.

After investigation, lines were found for the determinations of traces of terbium in dysprosium which showed no interference effects.

[25] H. V. Malmstadt and R. G. Scholz, *Anal. Chem.* 27, 881–83 (1955).

# MASS SPECTROMETRY

Russell Baldock

## METASTABLE AND COLLISION-INDUCED TRANSITIONS IN THE MASS SPECTRA OF *n*-BUTANE AND *i*-BUTANE

H. M. Rosenstock C. E. Melton

A careful study was made of metastable transitions in *n*-butane and *i*-butane, and further experimental tests were made of some aspects of the absolute rate theory of mass spectra.[1,2] Collision-induced dissociations corresponding to the spontaneous process

$$C_4H_{10}^{+} \overset{(\text{Collision})}{+} C_4H_{10} \longrightarrow C_3H_7^{+} + CH_3^{0}$$

were also observed, and some approximate cross sections were calculated. Since the limit of minimum detectability of these transitions was extended by one or two orders of magnitude, proper precautions were taken concerning instrumental sensitivity and stability, purity of gas samples, and low background current. The metastable transitions were distinguished from the collision-induced dissociations by pressure studies. If a metastable transition is truly a unimolecular process, the relative abundance should be independent of pressure, whereas a collision-induced process leads to a fragment whose relative abundance varies linearly with the pressure.

The behavior of a group of peaks in the mass range 13 to 18 is shown in Figs. 8 and 9. The 16.9 peak grows much more rapidly than the non-integral peaks at masses 13.85 and 15.3, indicating a collision process.

The relative abundance of masses 16.9, 13.8, and 12.6 is shown as a function of $C^+$ intensity in Fig. 10. It is also shown in Fig. 10 that a plot of relative abundance vs $C^+$ intensity leads to intercepts below the origin for data obtained in the higher pressure region. A study of the relation of $C^+$ ion intensity vs pressure indicated that the ion intensity is not proportional to pressure in this pressure range, thus explaining the deviation. The results are summarized in Tables 4 and 5.

Several of the collision-induced dissociations observed involve the loss of large neutral fragments. Nothing is known directly about the state of combination of these fragments after collision; however, the essentially linear pressure dependence of the relative abundance of these collision-induced processes requires that the decomposition occur as the result of a single collision. There is a striking parallel in the theories of collision-induced dissociation processes and the metastable processes resulting from electron excitation. That is, values for the apparent masses are obtained in both cases by the well-known equation for meta-

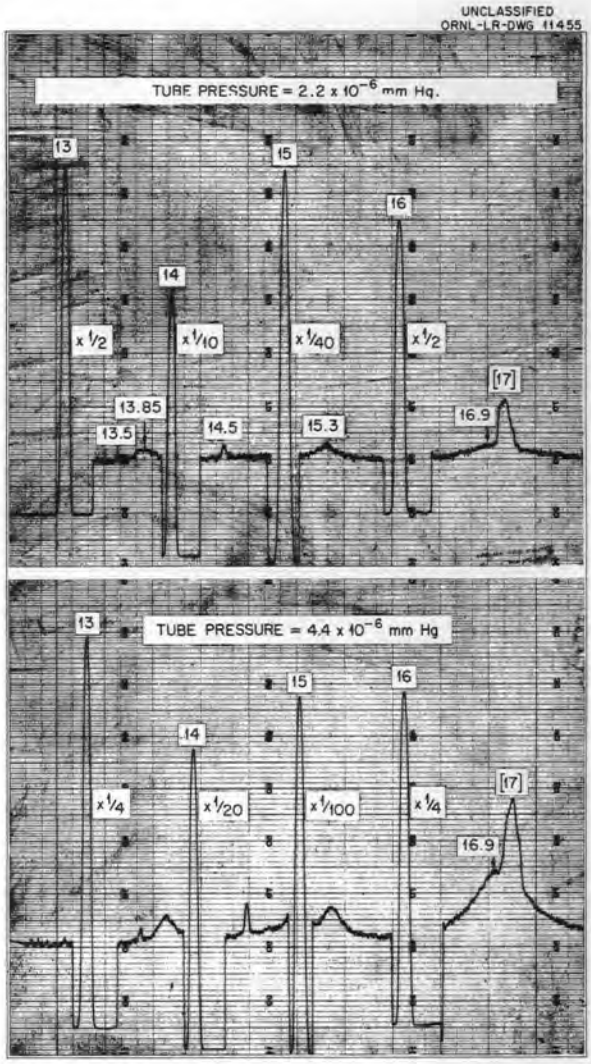



**Fig. 8. Metastable and Collision-induced Transitions of *n*-Butane.**

[1] M. B. Wallenstein, Ph.D. Thesis, University of Utah (1951).

[2] M. Wallenstein *et al.*, p 70–93 in *Symposium on Radiobiology* (J. J. Nickson, ed.), New York, 1952.

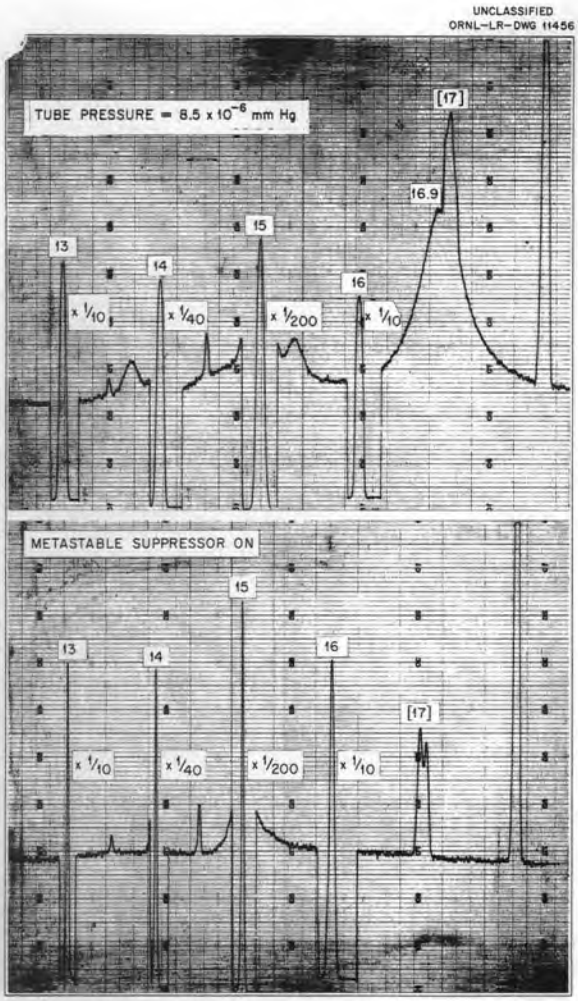



**Fig. 9. Metastable and Collision-induced Transitions of *n*-Butane with Metastable Suppressor On.**

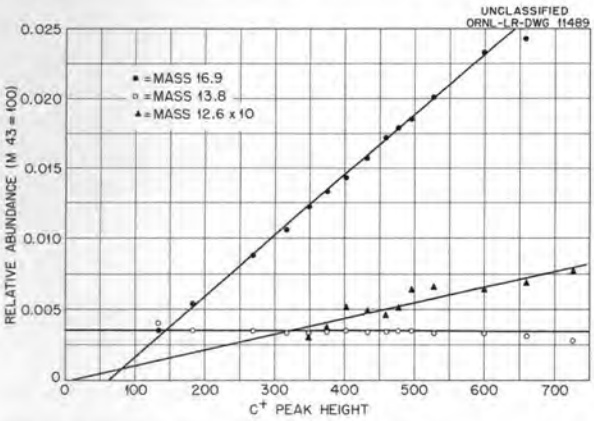



**Fig. 10. Relative Abundance of Masses 16.9, 13.8, and 12.6 as a Function of $C^+$ Intensity.**

stable transitions. These considerations lead to the suggestion that the only essential differences between the collision-induced reactions and the reactions induced by electron impact are the distribution of excitation energy and the detailed mechanism of excitation.

On the assumption that the collection efficiency of the various ions was the same, the relative cross sections given in Tables 4 and 5 were calculated. No dependable absolute-pressure measurements were possible in this work; however, rough calculations gave values of the order of $1 \times 10^{-16}$ cm$^2$ for the absolute cross section for the collision-induced reactions.

**TABLE 4. METASTABLE TRANSITIONS IN *n*-BUTANE AND *i*-BUTANE**

| Apparent Mass | Transition | Relative Abundance | |
|---|---|---|---|
| | | This Work | National Bureau of Standards |
| | | *n*-Butane | |
| 13.8 | 53 → 27 | 0.004 | |
| 15.3 | 55 → 29 | 0.004 | |
| 24.1 | 28 → 26 | 0.01 | 0.01 |
| 25.1 | 29 → 27 | 0.07 | 0.11 |
| 29.5 | 57 → 41 | 0.02 | |
| 30.4 | 58 → 42 | 0.08 | 0.14 |
| 31.3 | 59 → 43 | 0.001 | |
| 31.9 | 58 → 43 | 0.10 | 0.20 |
| 32.8 | 59 → 44 | 0.002 | |
| 35.1 | 39 → 37 | 0.007 | 0.02 |
| 37.1 | 41 → 39 | 0.04 | 0.06 |
| 39.1 | 43 → 41 | 0.34 | 0.46 |
| 43.0 | | (100.00) | (100.00) |
| | | *i*-Butane | |
| 13.8 | 53 → 27 | 0.003 | |
| 15.3 | 55 → 29 | 0.001 | |
| 24.1 | 28 → 26 | 0.005 | |
| 25.1 | 29 → 27 | 0.02 | 0.04 |
| 29.5 | 57 → 41 | 0.01 | |
| 30.4 | 58 → 42 | 0.015 | |
| 31.3 | 59 → 43 | 0.0007 | |
| 31.9 | 58 → 43 | 0.005 | |
| 35.1 | 39 → 37 | 0.01 | 0.02 |
| 37.1 | 41 → 39 | 0.05 | 0.08 |
| 39.1 | 43 → 41 | 0.52 | 0.72 |
| 43.0 | | (100.00) | (100.00) |

TABLE 5. COLLISION-INDUCED TRANSITIONS IN *n*-BUTANE AND *i*-BUTANE

Pressure: $1 \times 10^{-5}$ mm Hg

| Apparent Mass | Transition | Relative Abundance | Relative Cross Section | Abundance of Parent Ion |
|---|---|---|---|---|
| | | *n*-Butane | | |
| 12.8 | 57 → 27 | 0.0005 | (100) | 2 |
| 13.5 | 58 → 28 | 0.0005 | 20 | 9.5 |
| 14.5 | 58 → 29 | 0.0008 | 30 | 9.5 |
| 16.9 | 43 → 27 | 0.15 | 60 | 100 |
| 23.1 | 27 → 25 | 0.002 | 20 | 38 |
| 43.0 | | (100.00) | | |
| | | *i*-Butane | | |
| 16.9 | 43 → 27 | 0.03 | 1 | 100 |
| 23.1 | 27 → 25 | 0.0012 | 0.1 | 29 |
| 31.9 | 58 → 43 | 0.05 | (100.00) | 2 |
| 43.0 | | (100.00) | | |

Several collision-induced transitions were observed below mass 12. These transitions could not be assigned to specific fragments because of the difficulty in accurately determining their masses. A study of simple compounds such as $C_2H_2$, $CH_4$, and $C_2H_4$ would resolve this difficulty.

## ION BEAM ATTENUATION DUE TO CHARGE TRANSFER IN THE MASS SPECTROMETER

H. M. Rosenstock C. E. Melton

Difficulties were experienced in studies of collision phenomenon because of ion beam attenuation at pressures higher than those normally employed. A detailed investigation of this attenuation was carried out in argon to determine whether it was due to a space charge effect, elastic scattering, or charge transfer.

Data were taken in the pressure range $6 \times 10^{-8}$ to $6 \times 10^{-5}$ mm Hg. The $A^{36}$ ion intensity is shown as a function of the $A^{38}$ ion intensity in Fig. 11. Since the $A^{38}$ intensity is a constant fraction of the $A^{36}$ intensity, there is no mass discrimination in argon due to beam attenuation. The $A^{36}$ ion intensity as a function of pressure is shown in Fig. 12. The curve was fitted at a pressure of $4 \times 10^{-5}$ mm Hg to obtain the calculated values shown. If the ion beam attenuation shown in Fig. 12 is interpreted as charge transfer for the reaction

$$A^+ + A^\circ \longrightarrow A^\circ + A^+ ,$$

where $A^+$ is the argon ion and $A^\circ$ is neutral argon, the charge transfer cross section, $Q$, can be determined.

An average $Q$ value of 170 $cm^{-1}$ was calculated from several determinations. This value is a factor of 2 higher than that reported in the literature. The difference is probably due to an error in pressure measurements in this work. Pressure measurements were made in the vicinity of the cold trap, and the pressure in the analyzer tube could have been a factor of 2 higher than that measured by the ion gage. Since the beam attenuation observed closely follows a charge transfer type of reaction, it is suggested that the charge transfer is the predominant factor in ion beam attenuation in the mass spectrometer at the higher pressures.

## EXPERIMENTAL DETERMINATION OF ION BEAM WIDTH IN A MASS SPECTROMETER

J. R. Sites

The usefulness of a mass spectrometer is determined by its resolution, and the resolution depends on the radius of curvature of the deflected

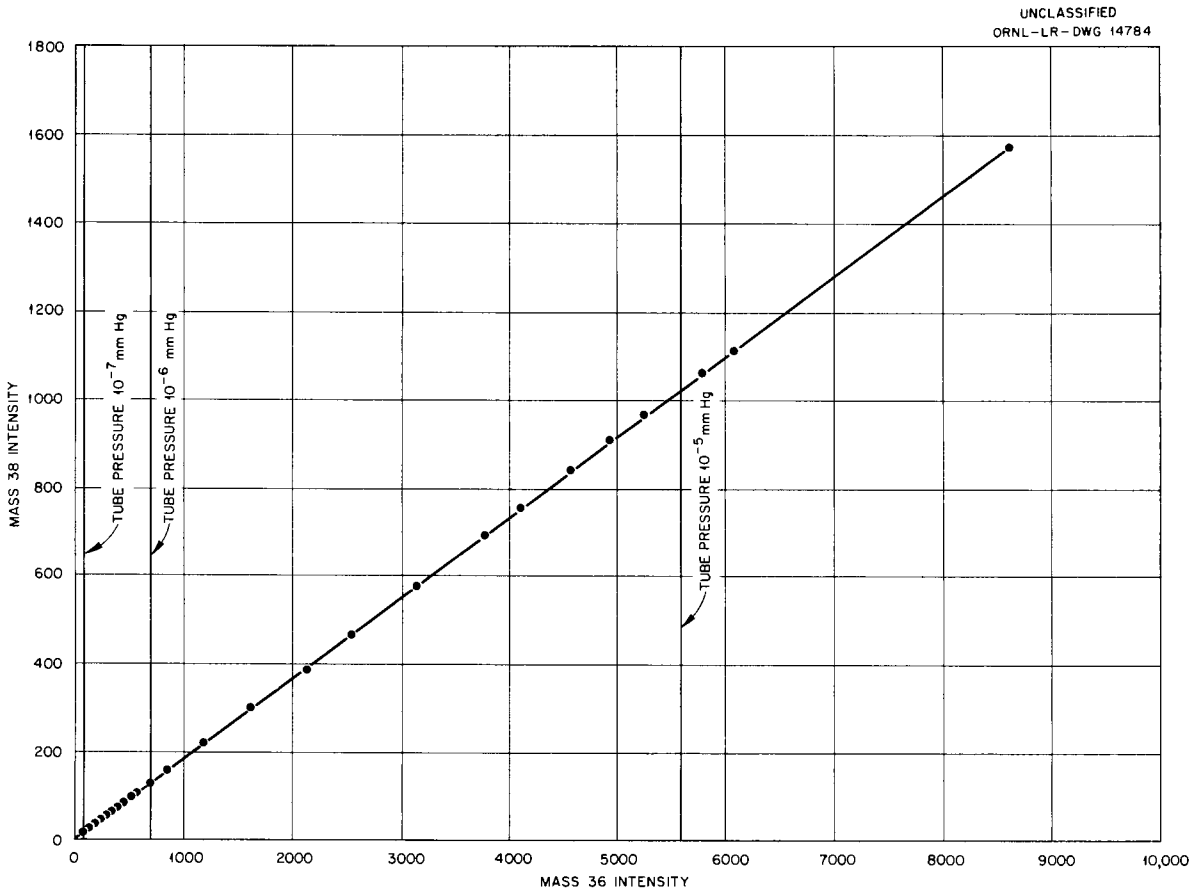



**Fig. 11. Plot of $A^{36}$ Ion Intensity vs $A^{38}$ Ion Intensity.**

ions, the width of the ion beam at the receiver, and the width of the receiver slit. The radius is determined by the dimensions of the spectrometer, and the receiver slit width can usually be varied by mechanical means. The beam width depends on the focusing potentials on the source slit system, the width of the final exit slit in the source, the maximum divergences of the ion beam from the source, the energy spread of the ions, and the alignment of the slits and the magnetic field.

It is desirable to have a sensitive and simple check of the beam width to be assured of optimum spectrometer conditions. For this, a relation between the beam width and simple measurements on the ion currents as displayed on recorder chart paper has been derived and checked.

Several assumptions have been made in order to simplify the derivation: (1) the ion intensity distribution across the ion beam is symmetrical and triangular; (2) there are two overlapping, adjacent ion peaks that are equal, or nearly equal; (3) the receiver slit width is greater than the beam width. The measurements to be made on the chart patterns should be sensitive to small changes in beam width, and should be made accurately with ease; that is, they should be determined by intersections of lines, sharp breaks, or flat-top peaks and not by estimating the middle of a broadened line profile.

The expression derived from the above considerations is

$$R_V = \frac{2}{1 + R_H}\left(1 - \frac{D}{B} + \frac{S_2}{B}\right) ,$$

where $B$ is the ion beam width, $D$ is the dispersion between the two masses being considered, $S_2$ is the width of the receiver slit opening, $R_V$ is the ratio of the valley between the two peaks to the height of the more intense peak, and $R_H$ is the ratio of the height of the more intense peak to that

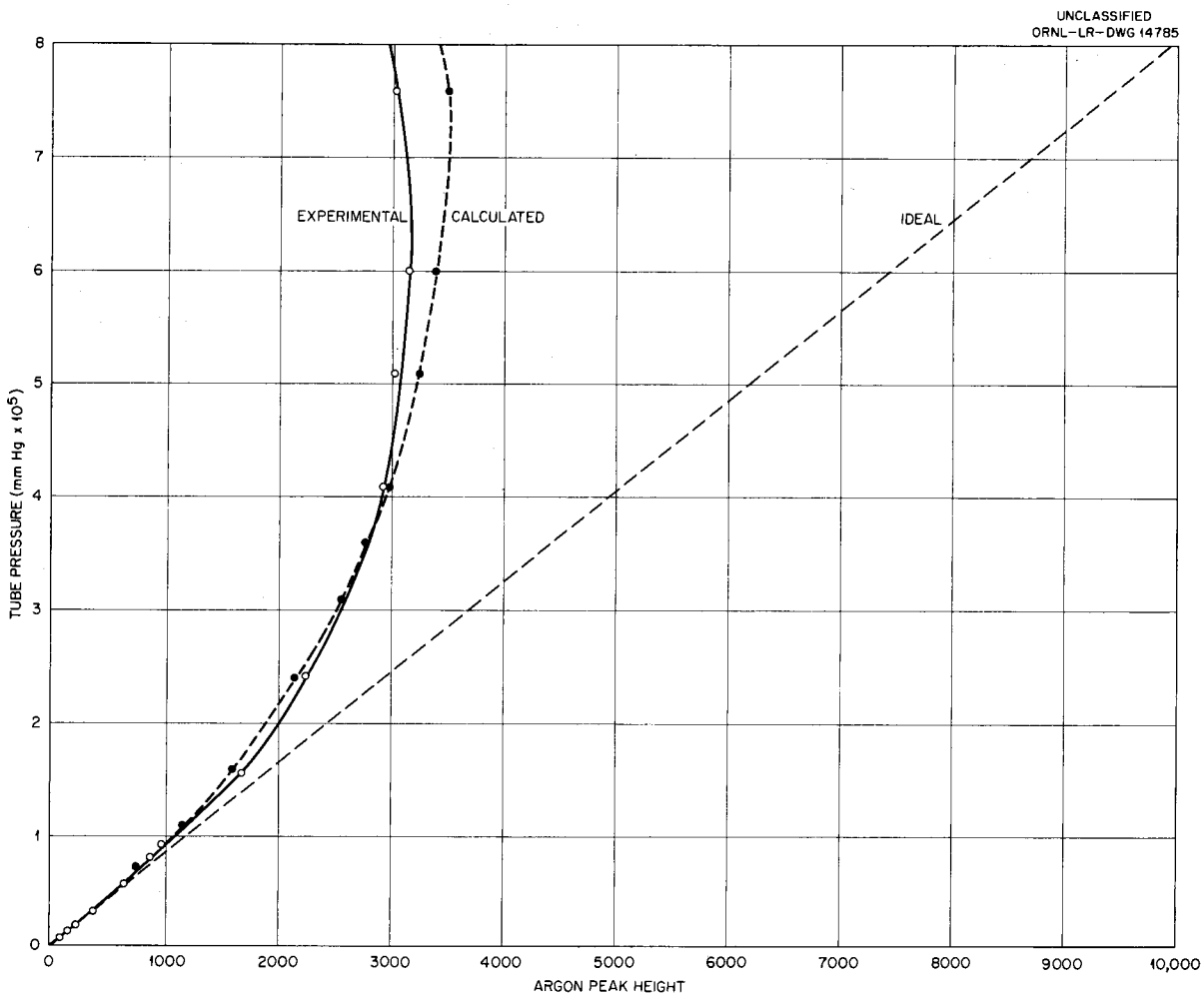



**Fig. 12. Argon Beam Attenuation at High Pressure.**

of the less intense peak. If the two peaks are equal, $R_H = 1$, the factor in front of the brackets becomes unity. As $S_2$ is varied, $R_V$ changes; and, since $D$ and $RH$ are constant for a given experiment, $B$ can be calculated.

If $R_V$ is plotted vs $S_2$, a straight line is obtained with a slope of $[2/(1 + R_H)]1/B$. The $S_2$-axis intercept, $R_V = 0$, gives $B = D - S_2$. From the slope, $B$ is determined independently of $D$ and $S_2$, which are subject to greater uncertainty than $R_H$. If $R_V = 2/(1 + R_H)$, $D = S_2$, which gives a check on the self-consistency of $R_H$, $D$, and $S_2$.

The beam width could also be determined by measurements of the widths of the base and the top of flat-topped, completely resolved peaks or from the expression $B = D - S_2$ – (width of flat base line between peaks). But these measurements require a very low rate of peak scanning to make the widths great enough to be measured accurately. On the other hand, the peak height and valley measurements required in the method presented here may be obtained with the scanning rates usually used for taking abundance data.

To test this method, a mixture of $U^{235}$ and $U^{236}$ has been made. Data are taken at the mass 267 and 268 positions of the $UO_2^+$ ions formed by surface ionization from a tantalum filament. For this material in the MS-A mass spectrometer, $D$ = 0.030 in., $R_H$ = 0.80, and $S_2$ was varied from its maximum of 0.043 in. to less than 0.001 in. The beam width determined from the slope of the $R_V$ vs $S_2$ plot is 0.014 in. Using the value for $S_2$ from

$R_V = 2/(1 + R_H)$, it is found that $B = 0.015$ in., a less reliable value.

According to a rule of thumb, the beam width is seldom less than twice the width of the source exit slit. In this case, the slit is 0.008 in., which gives 0.016 in. for the expected beam width. This agrees very well with the experimental values found.

Thus, for checking the resolution as adjustments are made, all that is needed is a plot of the ratio of the valley to the ion peak height for two adjacent overlapping ion peaks for a few receiver slit settings. The sensitivity is best in the range for $R_V$ of 0.2 to 0.6.

## STABLE ISOTOPE ANALYSES

J. R. Walton

Eighty-one calutron enriched isotope samples, received from the Stable Isotope Separation and Calutron Operations Department, were analyzed in mass spectrometer MS-C. These samples were composed of 20 different elements. The elements and compaunds used and the ions measured during the analyses are shown in Table 6.

TABLE 6. IONS MEASURED IN STABLE ISOTOPE ANALYSES

| Atomic No. | Element Investigated | Form Used | Oven | Ions Measured | |
|---|---|---|---|---|---|
| | | | | Thermal | Electron Produced |
| 5 | Boron | B | Ta | | $B^+$ |
| 12 | Magnesium | MgO | Ta | | $Mg^+$ |
| 16 | Sulfur | $As_2S_3$ | Ta | | $AsS^+$ |
| 20 | Calcium | $CaI_2$ | Ta | | $CaI^+$ |
| 22 | Titanium | $TiO_2$ | Ta | $TiO^+$ | |
| 28 | Nickel | NiO | Ta | | $Ni^+$ |
| 30 | Zinc | ZnO | Ta | | $Zn^+$ |
| 31 | Gallium | $Ga_2O_3$ | Ta | $Ga^+$ | |
| 32 | Germanium | $GeO_2$ | Ta | | $Ge^+$ |
| 37 | Rubidium | $RbNo_3$ | Ta | $Rb^+$ | |
| 38 | Strontium | $Sr(NO_3)_2$ | Ta | $Sr^+$ | |
| 42 | Molybdenum | $MoO_3$ | Ta | | $MoO^+$ |
| 44 | Ruthenium | Ru | Ir | $Ru^+$ | |
| 46 | Palladium | Pd | Ta | | $Pd^+$ |
| 47 | Silver | Ag | Ta | | $Ag^+$ |
| 52 | Tellurium | Te | Ta | | $Te^+$ |
| 56 | Barium | $BaCO_3$ | Ta | $Ba^+$ | |
| 60 | Neodymium | $Nd_2O_3$ | Ta | $NdO^+$ | |
| 62 | Samarium | $Sm_2O_3$ | Ta | $Sm^+$ | |
| 63 | Europium | $Eu_2O_3$ | Ta | $Eu^+$ | |
| 74 | Tungsten | $WO_3$ | Ta | | $WO^+$ |
| 78 | Platinum | Pt | Ta | | $Pt^+$ |

### TWO-STAGE MASS SPECTROMETER MS-F

G. F. Wells Russell Baldock

The two main magnets and bed plate for mass spectrometer MS-F have been fabricated and are now being inspected prior to installation.

Design is complete on the main tubes, source, receiver, and first focal point sections, which are shown assembled in Fig. 13. The entire system is to be wrapped with a Nichrome strip heating system to provide high temperature bake-out. Cross bracing of the individual tubes is rigid to maintain maximum stability of the relationship between foci for each tube. The vertical supports for the second tube have a weaker bending moment than those for the first tube. This allows it to move relative to the first on thermal expansion between the two tubes. Other features are as described previously,[3] with the exception of the source isolation valve, shown in Fig. 14. This valve has been reoriented to seat against source chamber pressure instead of with it. This allows the final exit slit of the source to seat against the valve aperture on the source chamber side.

The only opening between this chamber and the tube is the 0.250 × 0.010 in. source exit slit itself. This will allow differential pumping between the two sections, and the tube can be maintained at a lower pressure than the pressure in the source chamber. Such an arrangement is in accord with the effort toward achieving the best vacuum obtainable in the tubes.

[3]G. F. Wells and J. R. Sites, *Stable Isotopes Research and Production Semiann. Prog. Rep. Nov. 20, 1955*, ORNL-2028, p 18.

### MASS SPECTROMETER ASSAY

L. O. Gilpatrick C. E. Prather
C. E. Melton J. R. Walton

Spectrometer assays for November through April amounted to 858, an increase of 17% over the previous six months. The breakdown within the Laboratory is shown in Table 7.

TABLE 7. MASS SPECTROMETER ASSAYS, NOVEMBER 1955–APRIL 1956

| Division or Project | Total Number of Samples |
|---|---|
| Stable Isotopes Division | 201 |
| Materials Chemistry Division | 293 |
| Chemistry Division | 158 |
| Metallurgy Division | 6 |
| Reactor Projects | 133 |
| Miscellaneous | 67 |

UNCLASSIFIED
ORNL-LR-DWG 13671

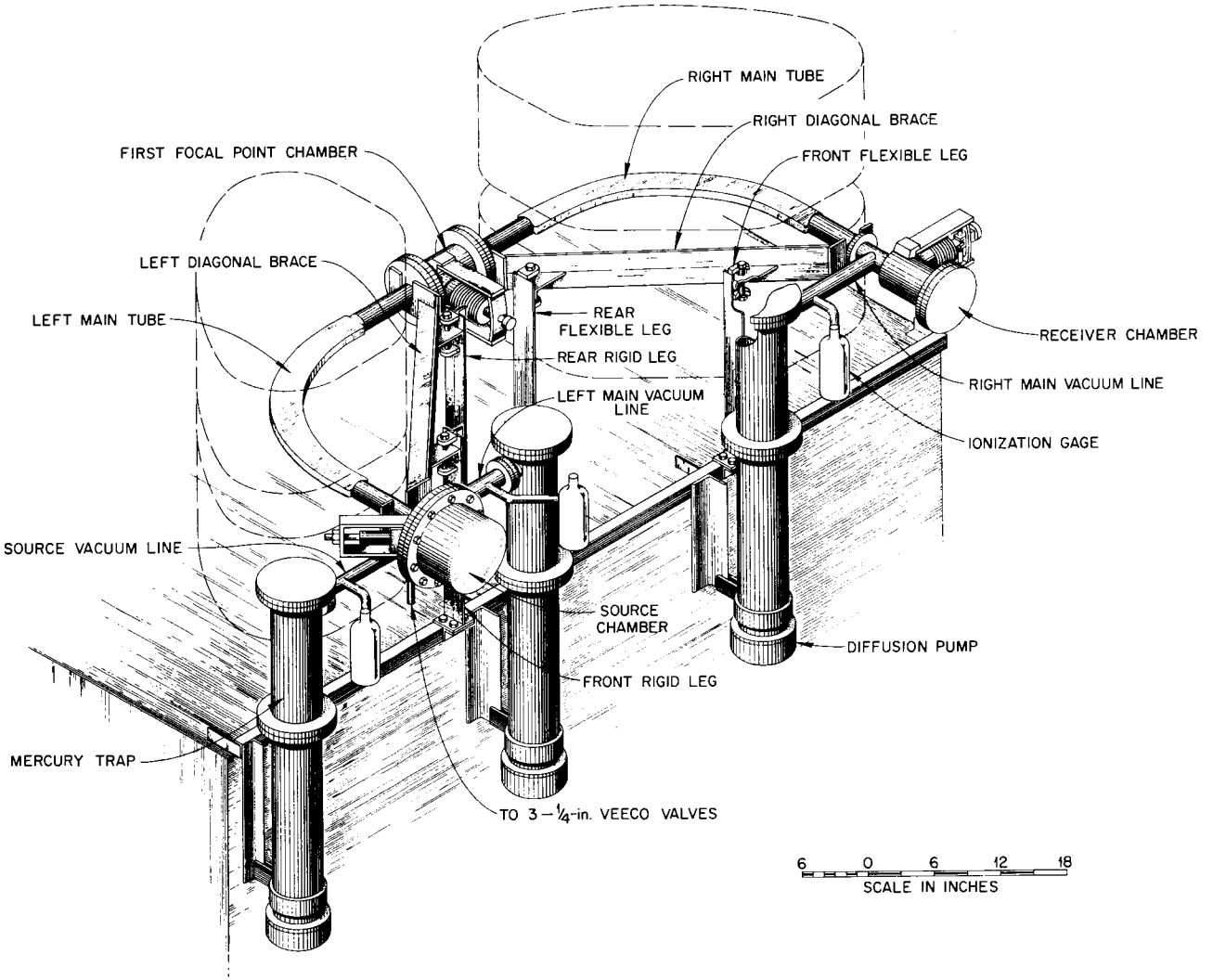



**Fig. 13. Two-Stage Mass Spectrometer Tubes and Vacuum System.**

UNCLASSIFIED
ORNL-LR-DWG 13672

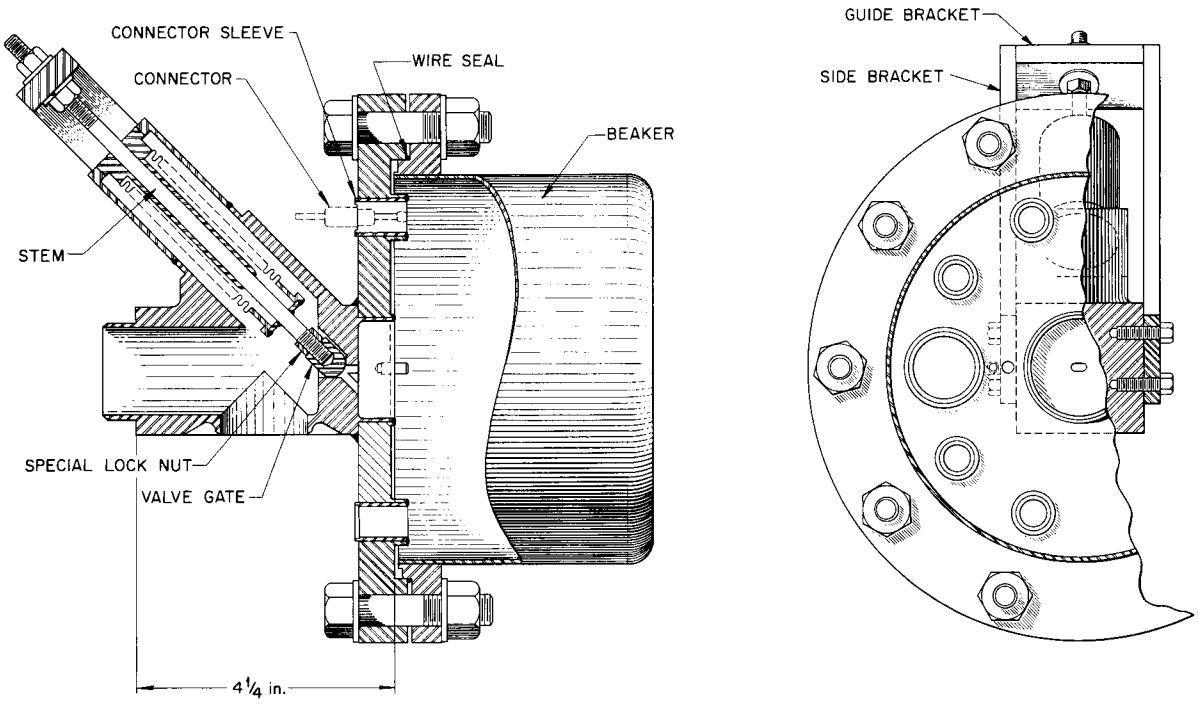



**Fig. 14. Two-Stage Mass Spectrometer Source Isolation Valve.**

# STABLE ISOTOPE SEPARATION AND CALUTRON OPERATIONS

A. E. Cameron

## INTRODUCTION

Replenishment of the inventory has been the principal activity of the electromagnetic separation work during the past six months. The over-all operation has been uniformly satisfactory. The spasmodic difficulty experienced with one Beta tank during a separation run on magnesium in early December has not been satisfactorily explained. In all the separations made the emphasis has been on product purity in an attempt to comply with customer demands for higher enrichments. To some extent it is possible to meet such needs without making a second pass, which would require the accumulation of an inventory of pre-enriched feed material. Improvements in collector pockets and in baffling techniques have contributed to the success of such runs. In some instances source operation at other than maximum obtainable ion output was necessary.

The elements processed include, in addition to completion of barium,[1] Mg, W, Ag, Ge, Cd, Zn, Eu, Tl, Cu, Rb, and Re; Cl and Pb are currently in process. Thus, an aggregate of 51 isotopes of 14 elements has been processed in this six-month interval. Integrated ion currents at the collectors indicate the total production to be 975 g.

## CALUTRON OPERATIONS

| | |
|---|---|
| L. O. Love | K. A. Spainhour |
| W. A. Bell, Jr. | C. V. Ketron |
| G. M. Banic, Jr. | R. L. Caldwell |
| W. K. Prater | W. W. Davis |
| C. W. Sheridon | G. J. Fisher |

### Barium, Series HJ-XAX[2]

Collection of barium isotopes ($Ba^{130}$, $Ba^{132}$, $Ba^{134}$, $Ba^{135}$, $Ba^{136}$, $Ba^{137}$, $Ba^{138}$) was started on September 22 and continued through December 3, 1955. Comparison of operability of $BaCl_2$ and metallic barium charges containing strontium contaminants varying from 1 to 4% was reported previously.[1] Collections performed since those reported have been made only with metallic barium charges containing 1% strontium contaminant. Average current at the collector during use of this charge was 21.8 ma per hour of ion reception in comparison with an average of 6.1 ma/hr when the charge contained from 2 to 4% strontium.

The following estimated amounts were collected:

| Isotope | Amount (g) |
|---|---|
| $Ba^{130}$ | 0.3 |
| $Ba^{132}$ | 0.5 |
| $Ba^{134}$ | 3.4 |
| $Ba^{135}$ | 11.0 |
| $Ba^{136}$ | 15.2 |
| $Ba^{137}$ | 23.7 |
| $Ba^{138}$ | 117.0 |
| Total | 171.1 |

Of the total amount, 108 g was collected in the interim September 22, 1955, to November 20, 1955.

### Magnesium, Series HM-XBX

Separation of magnesium isotopes ($Mg^{24}$, $Mg^{25}$, $Mg^{26}$) was started on November 6 and was completed on December 23, 1955. During this collection a comparison was made between operation of an ion source utilizing a single arc and an identical type of source utilizing two arcs. Metallic magnesium was used as charge feed. Separated ions were collected in water-cooled copper pockets placed behind beam-defining slots, each separated from the next adjacent slot by 1.004 in. and each having an opening of 0.187 in. All pockets contained a substantial quantity of copper oxide wire upon which the magnesium ion beams impinged, possibly reacting to form some magnesium oxide in the pocket. The formation of such a compound could materially aid in increasing the amount of incidental material that formerly had been rejected by sputtering and vaporization (as much as 90% in the most extreme case). The retention and purity of the material produced in this operation are compared, in Table 8, with those of the material produced in the previous collection performed in 1949. Improvement in both retention and isotopic purity is noted in the present collection.

---

[1] L. O. Love, *Stable Isotopes Research and Production Semiann. Prog. Rep. Nov. 20, 1955*, ORNL-2028, p 24 ff.

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

TABLE 8. COMPARISON OF ISOTOPIC RETENTION AND PURITY OF MAGNESIUM

| Isotope | Natural Abundance (%) | Enriched Abundance (%) | | Retention of Material (%) | |
|---|---|---|---|---|---|
| | | 1949 | 1955 | 1949 | 1955 |
| $Mg^{24}$ | 78.60 | 99.59 | | 9.3 | 60.3 |
| $Mg^{25}$ | 10.11 | 92.33 | 97.5 | 31.3 | 58.3 |
| $Mg^{26}$ | 11.29 | 98.12 | 98.99 | 18.9 | 63.0 |

Source operation with one arc was considerably smoother than that encountered with two arcs. The two-arc unit sparked almost continuously and tended toward overheating from varying drain conditions; also, negative-electrode support-bushing failures were encountered. The peak output obtained from the two-arc source was approximately twice that obtained from single-arc operation; however, the average outputs per hour of ion reception were nearly equal. Other parameters of operation are compared for the two units in Table 9.

The following estimated amounts of each isotope were collected:

| Isotope | Amount (g) |
|---|---|
| $Mg^{24}$ | 87.8 |
| $Mg^{25}$ | 9.6 |
| $Mg^{26}$ | 10.0 |
| Total | 107.4 |

### Tungsten, Series HN-XAX

This collection of the isotopes of tungsten ($W^{180}$, $W^{182}$, $W^{183}$, $W^{184}$, $W^{186}$) was started on December 20, 1955, and continued through January 12, 1956. The charge feed was $WCl_6$ used in a low-temperature ion source operating in the 75 to 125°C range. The ions produced were collected in water-cooled copper receiving pockets positioned behind the slots of a graphite beam-defining plate. Each of these slot openings was 0.125 in. in width, with their center lines being separated by 0.276 in. for collection of isotopes whose masses differ by one mass unit. This spacing was so close that the adjacent pockets had to be electrically insulated with thin mica spacers, which soon became coated with material rejected from the pocket. Correct monitoring of the beams was then quite difficult. The major source problem was that caused by deposition of material around the inside periphery of the electron collimating slot. With continued growth in this location, electrons from the filament were blocked from entering the ionization chamber, and run termination was inevitable.

The total time involved in this collection was 1837 hr, during which time ion reception was maintained for 501 hr. The average ion collection rate was 10.5 ma per hour of reception, with a process efficiency of 4.1%.

The following estimated amounts of tungsten isotopes were collected:

| Isotope | Amount (g) |
|---|---|
| $W^{180}$ | 0.7 |
| $W^{182}$ | 20.9 |
| $W^{183}$ | 14.9 |
| $W^{184}$ | 23.9 |
| $W^{186}$ | 24.7 |
| Total | 85.1 |

### Silver, Series HO-XBX

This separation of the isotopes of silver ($Ag^{107}$, $Ag^{109}$) was started on December 23, 1955, and continued through January 11, 1956. An AgCl charge, operating in excess of 650°C, was used in the graphite-heater, type M-14 ion source, and nitrogen was used to support the silver arc. Cold surfaces of the source, particularly the accelerating electrodes, were coated with heavy deposits of silver soon after operation was started. Sparking around the source was severe, and the deposits were frequently melted, thus forming protrusions on the electrodes which resulted in distortion of the ion beam at the receiver.

The produced ions were received in water-cooled copper collector pockets positioned behind defining slots each 0.125 in. in width. Pocket center lines were separated by 0.465 in.

An excessive amount of copper and silver was rejected from the collector pockets. This material condensed on the back of the defining face-slots and built up until the pocket became electrically shorted to ground. In an effort to minimize this rejection of valuable isotopes, the bottom of the $Ag^{107}$ pocket was effectively lowered out of the path of the beam by utilizing a larger pocket and

TABLE 9. OPERATING PARAMETERS FOR SINGLE- AND TWO-ARC ION SOURCES – MAGNESIUM SEPARATION

| Source | Arc | Maximum Output (ma) | Average Output per Hour (ma) | | Process Efficiency (%) | Isotopic Purity (%) | |
|---|---|---|---|---|---|---|---|
| | | | During Ion Reception Time | During Total Tank Time | | $Mg^{25}$ | $Mg^{26}$ |
| Single | Single | 265 | 191 | 131 | 9.4 | 97.5 | 98.99 |
| | Upper | 227 | 105 | 53 | | | 96.7 |
| Two-Arc | Lower | 210 | 108 | 54 | | 93.1 | |
| | Total | 437 | 213 | 107 | 6.6 | | |

by lining the bottom of the $Ag^{109}$ pocket, which could not be treated in like manner, with a sheet of graphite. The isotopic material deposited on the graphite was in the form of very hard black flakes almost wholly impervious to acid attack.

A total of 891 tank hours was used in this separation. Ion reception was maintained for 471 hr at an average rate of 45.5 ma, peak output obtained was 70 ma, and process efficiency was 6.1%.

The following estimated amounts of silver isotopes were collected:

| Isotope | Amount (g) |
|---|---|
| $Ag^{107}$ | 69.9 |
| $Ag^{109}$ | 65.9 |
| Total | 135.8 |

### Germanium, Series HP-XBX

The separation of the isotopes of germanium ($Ge^{70}$, $Ge^{72}$, $Ge^{73}$, $Ge^{74}$, $Ge^{76}$) was performed in the interim of January 11 to January 30, 1956. A $GeCl_4$ charge was fed into the ionization chamber from a flask, external to the vacuum chamber. Rate of charge feed then was regulated by a conventional leak valve. The ions produced were collected in graphite receiving pockets. Each pocket was positioned behind a 0.125-in. beam-defining slot, and the separation between pocket centers was 0.347 in. per mass unit. Pockets for the reception of $Ge^{72}$, $Ge^{73}$, and $Ge^{74}$ were fit snugly against the beam-defining faceplate to form a shield between the other isotope collectors. Only the ion currents in the $Ge^{70}$ and $Ge^{76}$ pockets were monitored. Amounts collected in the other three pockets then were estimated on the basis of natural abundance ratios. The germanium in the calutron system not actually deposited in the receiver pockets was recovered for use as future charge material.

This separation required the use of 451 tank hours. Ion reception was achieved during 394 hr, the average rate of collection being 42.4 ma. Peak reception was 68.2 ma, and efficiency of the process was 12.3%.

The following estimated amounts of germanium isotopes were collected:

| Isotope | Amount (g) |
|---|---|
| $Ge^{70}$ | 9.5 |
| $Ge^{72}$ | 12.3 |
| $Ge^{73}$ | 3.5 |
| $Ge^{74}$ | 16.9 |
| $Ge^{76}$ | 3.1 |
| Total | 45.3 |

### Cadmium, Series HQ-XAX

The separation of cadmium isotopes ($Cd^{106}$, $Cd^{108}$, $Cd^{110}$, $Cd^{111}$, $Cd^{112}$, $Cd^{113}$, $Cd^{114}$, $Cd^{116}$) was performed in the interval January 10 to February 18, 1956. Maximum emphasis was placed on the attainment of high isotopic purity and less on the production of ions at the maximum obtainable rate. Ions were produced from $CdCl_2$ charge vaporized into the ionization chamber of a medium-temperature ion source. These ions were collected in water-cooled copper pockets. Ion entry into each pocket was through a 0.125-in.

beam-defining slot, with the separation per mass unit between slots being 0.450 in. To prevent the diffusion of rejected material from one pocket to another, the $Cd^{111}$ and $Cd^{113}$ collectors were fitted snugly against the beam-defining faceplate. In like manner, formed graphite strips were fitted against the defining faceplate between the $Cd^{106}$ and $Cd^{108}$ and between the $Cd^{108}$ and $Cd^{110}$ pockets. The amount of material collected in the $Cd^{111}$ and $Cd^{113}$ pockets was estimated on the basis of natural abundance ratios in comparison with monitored currents received in the other collectors.

A total of 1692 separator-hours was devoted to this cadmium separation. Ion reception was maintained for 1063 hr, during which period the total isotope beam current at the collector averaged 24.3 ma. The peak ion current attained was 51.9 ma, and process efficiency was 6.6%.

The following estimated amounts of the isotopes of cadmium were collected:

| Isotope | Amount (g) |
|---|---|
| $Cd^{106}$ | 1.0 |
| $Cd^{108}$ | 1.0 |
| $Cd^{110}$ | 12.6 |
| $Cd^{111}$ | 13.5 |
| $Cd^{112}$ | 25.6 |
| $Cd^{113}$ | 13.2 |
| $Cd^{114}$ | 31.5 |
| $Cd^{116}$ | 8.6 |
| Total | 107.0 |

### Zinc, Series HR-XBX

This separation was started on January 30, 1956, to replenish the inventory of the isotopes of zinc ($Zn^{64}$, $Zn^{66}$, $Zn^{67}$, $Zn^{68}$, $Zn^{70}$) and was completed on February 13.

The operation was performed in a medium-temperature ion source in which metallic (mossy) zinc charge was vaporized into the arc chamber. Ions were produced in an arc supported with nitrogen gas. Isotopic material was collected in water-cooled copper receiving pockets, each pocket being positioned behind a 0.125-in. beam-defining slot. Pockets for the collection of $Zn^{66}$ and $Zn^{68}$ were fitted snugly against the beam-defining faceplate to minimize the diffusion of material rejected from one pocket into an adjacent collector. Only ion beams of $Zn^{64}$, $Zn^{67}$, and $Zn^{70}$ were electrically monitored, and the amount of material collected in the $Zn^{66}$ and $Zn^{68}$ pockets was estimated on the basis of natural abundance ratios.

A total of 669 separator-hours was devoted to the collection. Metered ion reception was maintained for 526 hr at an average rate of 70 ma, the peak production being 139.4 ma. Efficiency of the process was 10.3%.

The following estimated amounts of isotopes were collected:

| Isotope | Amount (g) |
|---|---|
| $Zn^{64}$ | 43.0 |
| $Zn^{66}$ | 25.3 |
| $Zn^{67}$ | 3.7 |
| $Zn^{68}$ | 17.4 |
| $Zn^{70}$ | 0.5 |
| Total | 89.9 |

### Europium, Series HS-XAX

This separation was made in order to produce approximately 500 mg of high-purity $Eu^{153}$. The operation, which consisted of only one run, was started on February 5 and ended on February 10, 1956. During this interim the tank was cleaned thoroughly, and upon termination of the operation all europium in the system was salvaged for recycling into future $EuCl_3$ charge material.

The separated isotopic material was collected on purified graphite inserts. The insert upon which $Eu^{153}$ impinged was contained in a graphite receiving pocket, whereas the insert for reception of $Eu^{151}$ was contained in a water-cooled copper pocket. These inserts covered only one-third the entire pocket area, and the material collected thereon was sampled separately from that in the pocket proper. The $Eu^{151}$ pocket was made of copper in order to condense as much as possible of the $Eu^{151}$ rejected from the receiving insert and thus minimize contamination of the $Eu^{153}$ sample. A further precaution taken to minimize this type of contamination included positioning of the $Eu^{153}$ pocket snugly against the beam-defining faceplate. Such an arrangement permitted monitoring of the $Eu^{151}$ ion beam only, and the amount of $Eu^{153}$ collected was estimated on the basis of natural abundances.

The purity of material collected on the $Eu^{153}$ insert was 99.4% in comparison with 95.0% $Eu^{153}$ produced previously.

A total of 104 separator-hours was devoted to the collection. Ion collection was maintained for 31 hr at an average rate of 7.3 ma, the maximum reception attained being 13.2 ma. Efficiency of the process was 5.6%.

The following estimated amounts of europium isotopes were collected:

| Isotope | Amount (g) |
|---|---|
| $Eu^{151}$ | 0.6 |
| $Eu^{153}$ | 0.7 |
| Total | 1.3 |

### Thallium, Series HT-XAX

The separation of thallium isotopes ($Tl^{203}$, $Tl^{205}$) was performed in the interim of February 17 to March 19, 1956. Ions were produced from TlI charge in a low-temperature type of ion source. The separated material was collected in water-cooled copper pockets, with the ion beam entering each pocket through a 0.125-in. defining slot. The $Tl^{205}$ pocket was fitted against the beam-defining faceplate as a shielding technique.

During operation, severe collector pocket erosion was encountered, with large amounts of copper and thallium condensing on the cold surfaces of the beam-defining faceplate. When sufficient material was deposited to electrically short out the pocket, termination of the run became mandatory. Graphite liners were placed in the bottom of the pockets to retard pocket erosion and sputtering, but their effectiveness was questionable. Copper sputtered off the top and base of the pocket, coating the graphite and forming a copper surface which could be subjected to but very little cooling. Retention of isotopic material in these two instances is being determined.

A total of 1470 separator-hours was devoted to the series. Ion reception was maintained for 1157 hr at an average rate of 18.1 ma, the maximum rate being 44.0 ma.

The following estimated amounts of isotopic material were collected:

| Isotope | Amount (g) |
|---|---|
| $Tl^{203}$ | 46.2 |
| $Tl^{205}$ | 113.9 |
| Total | 160.1 |

### Copper, Series HV-XBX

Separation of the isotopes of copper ($Cu^{63}$, $Cu^{65}$) was performed in the interval March 15 to April 11, 1956, with the use of a CuCl charge in a medium-temperature type of ion source. Ion collection was in purified graphite pockets, with the ion beam entering each pocket through a 0.125-in. defining slot. The $Cu^{65}$ pocket was fitted against the beam-defining faceplate as a shielding technique. Collected amounts of $Cu^{65}$ were estimated from the monitored $Cu^{63}$ ion beam current on the basis of natural abundance ratios.

The $Cu^{63}$ collector pockets were changed frequently, and each pocket (or in one instance a group of pockets) was processed individually to determine retention of copper and isotopic purity as functions of, respectively, production rate with a constant amount collected in each pocket and amount of copper collected, with the output held as a constant. Product purity resulting from the use of an ion-exit slit reduced to one-half standard length was to be compared with that produced at a similar rate by a conventional ion source. These data are plotted in Figs. 15 and 16. Significant trends in retention and purity are lacking except for the increase in sample purity attained with the use of a shortened ion-exit slit.

A total of 1252 separator-hours was devoted to the series. Ion reception, as metered at the collector, was maintained for 1005 hr at an average rate of 22 ma. Ion output in each run was held at

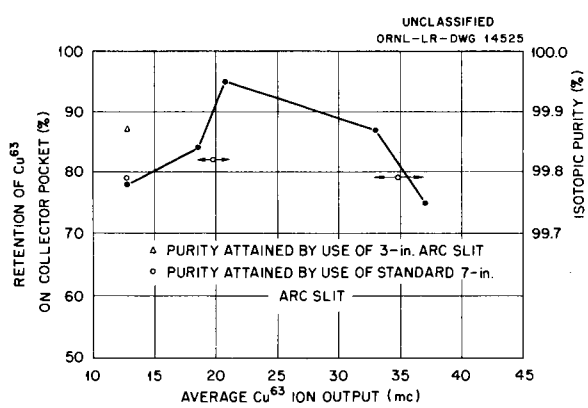



**Fig. 15. Retention of $Cu^{63}$ vs Average $Cu^{63}$ Ion Current.** Estimated amount of $Cu^{63}$ collected in each pocket, 2.1 g.

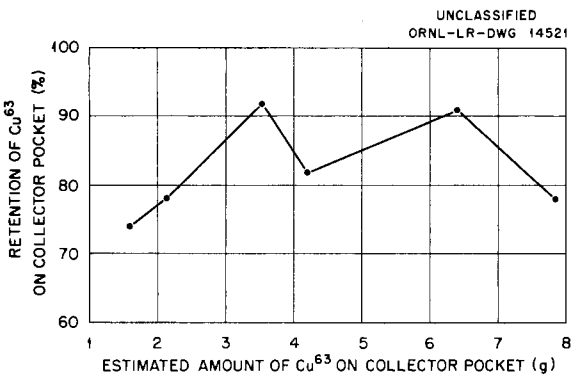



**Fig. 16. Retention of $Cu^{63}$ vs Estimated Amount of Collection.** Output of ions held constant at 12 to 14 ma.

predetermined values, the maximum attained being 81 ma. Process efficiency was 5.5%.

The following estimated amounts of copper isotopes were collected:

| Isotope | Amount (g) |
|---|---|
| $Cu^{63}$ | 36.4 |
| $Cu^{65}$ | 16.0 |
| Total | 52.4 |

### Rubidium, Series HW-XAX

Separation of the isotopes of rubidium ($Rb^{85}$, $Rb^{87}$) was performed from March 19 to April 26, 1956. High-purity isotopes were desired; hence operations were planned to obtain optimum production rate. Focus of the ion beams was of prime importance, and a recycle of RbCl into future charge material was necessary.

The ion beams produced were collected in copper pockets, each having extensive means for water-cooling both the top and bottom of the pocket. Prior rubidium separations had proved that erosion of conventional receiver pockets was extremely severe. The additional cooling applied to these pockets retarded the erosion to the extent that it was not a major cause of run termination in this series of collections. The ion beam entered each pocket through a 0.125-in. defining slot separated from the next adjacent slot by a distance of 1.66 in. This separation between the two ion beams permitted a graphite box to be placed around the $Rb^{87}$ pocket to serve as a shield against contamination by un-ionized material.

The total separator time involved in this collection was 1826 hr. Ion reception was maintained for 1272 hr at an average rate of 3.6 ma, the peak reception being 6.6 ma. Process efficiency was 9.5%.

The following estimated amounts of rubidium isotopes were collected:

| Isotope | Amount (g) |
|---|---|
| $Rb^{85}$ | 10.8 |
| $Rb^{87}$ | 3.9 |
| Total | 14.7 |

### Rhenium, Series HX-XBX

Separation of the two isotopes of rhenium ($Re^{185}$, $Re^{187}$) was started on April 11 and was completed on April 22, 1956, the length of time being dependent upon the availability of rhenium and the time required to recycle the material into fresh charges of $Re_2O_7$.

During operation the $Re_2O_7$ charge apparently decomposed, and constant increases in oven temperature were required to hold ion production constant. On termination of the runs, charge residues were no longer yellow but were either purple or black, with black residues left in the copper castings of the ion source. Change in charge composition with time is further evidenced by the change in the number and intensity of extraneous sideband ion beams encountered during operation.

The ions produced were collected in graphite pockets, each placed behind a $\frac{1}{8}$-in. beam-defining slot. Beam separation at the receiver being only 0.271 in. made it mandatory that the two pockets be in contact with each other. Thus, ion current to both receiving pockets was monitored on a single meter, and amounts of isotopes collected were estimated from natural abundances. The rhenium collected in the pocket was deposited in the form of hard, brittle flakes. There was but little evidence of pocket erosion; however, the top of each pocket had warped badly and was cracked in two or three places.

The total separator time involved in this separation was 502 hr. Ions were received in the collectors for 339 hr at an average rate of 3.3 ma, the maximum rate attained being 14.4 ma. Process efficiency was only 1.2%, primarily because of the formation of extraneous ions within the arc.

The following estimated amounts of rhenium isotopes were collected:

| Isotope | Amount (g) |
|---|---|
| $Re^{185}$ | 3.2 |
| $Re^{187}$ | 4.7 |
| Total | 7.9 |

### Chlorine, Series HY-XBX

Separation of the isotopes of chlorine ($Cl^{35}$, $Cl^{37}$) was started on April 20, 1956, and is nearing completion. The separated material is retained by chemical combination with either magnesium chips or lead pellets. Preliminary material recoveries and mass analyses indicate that each element performs about equally well as a means of chlorine collection. The charge material being used in this separation is $FeCl_2$, instead of $Cl_2$, to reduce the possibility of $MgCl_2$ formation by molecular chlorine.

The following estimated amounts of chlorine isotopes have been collected:

| Isotope | Amount (g) |
|---|---|
| $Cl^{35}$ | 19.8 |
| $Cl^{37}$ | 6.2 |
| Total | 26.0 |

### Lead, Series HZ-XAX

The separation of the four isotopes of lead ($Pb^{204}$, $Pb^{206}$, $Pb^{207}$, $Pb^{208}$) was started on April 26, 1956, and is nearing completion. High isotopic purity is of first importance, and ion output has not been pushed to its maximum value.

The separation (0.245 in.) between adjacent beams at the receiver makes desirable the omission of alternate pockets; thus $Pb^{204}$, $Pb^{206}$, $Pb^{208}$ are collected in one receiver and $Pb^{204}$ and $Pb^{207}$ are collected in the other. A receiver designed to collect all isotopes simultaneously has not been completely successful because of pocket erosion and consequent sample contamination.

The following estimated amounts of lead isotopes have been collected to date:

| Isotope | Amount (g) |
|---|---|
| $Pb^{204}$ | 2.0 |
| $Pb^{206}$ | 17.7 |
| $Pb^{207}$ | 18.9 |
| $Pb^{208}$ | 37.0 |
| Total | 75.6 |

## PREPARATION OF CHARGE MATERIALS

C. W. Sheridan

The quality of charge materials used in calutron operations contributes immensely to the success of isotope separations. Moisture in a charge is particularly objectionable, and a procedure for dehydration at approximately 400°C at reduced pressure has become a standard practice.

Of the 14 different charge materials (Ba, Mg, $WCl_6$, AgCl, $GeCl_4$, $CdCl_2$, Zn, $EuCl_3$, TlI, CuCl, RbCl, $Re_2O_7$, $FeCl_2$, $PbCl_2$) used in the past six months, only 6 (Mg, $GeCl_4$, $CdCl_2$, Zn, CuCl, $PbCl_2$) were purchased as such, and these (except germanium) were subjected to the above outgassing procedure before they were installed in the calutron. Metallic barium was purified by fractional distillation to remove the strontium which was present in the commercial product. All other charges were synthesized in the laboratory, using dry techniques whenever possible.

Recycle of the charges of germanium and europium has been completed, and recycle of rubidium and rhenium is in progress. Germanium from the calutron and from the vacuum exhaust gases (scrubbed with NaOH) was processed in 6 *N* sulfuric acid followed by $H_2S$ precipitation. Subsequent treatment with concentrated $NH_4OH$ followed by $H_2O_2$ yielded $GeO_2$. Losses in the calutron system were severe, only 60% of the germanium installed in the system being recovered. The loss may have been in the pump oils, which were not processed; or the scrubbing of exhaust gases may have been ineffective, as little germanium was found in the scrubber system.

The recycle of europium consisted in washing calutron parts, filtering the solutions, precipitating the europium as the hydroxide and oxalate, and finally igniting the oxalate to $Eu_2O_3$. Approximately 80% of the material supplied as charges has been recovered in this manner.

Rubidium from the calutron is being recovered by precipitating the impurities with $NH_4OH$, followed by $H_2S$. Subsequent chlorination produces future RbCl charge. Calutron losses in the process are expected to be about 15%.

The recycle of rhenium is nearing completion. Washings from the calutron are put into solution as sodium perrhenate. The solution is filtered, the filtrate taken to dryness, then again filtered from a solution having a pH of 8. The solution is acidified to 10% by weight with HCl, and rhenium sulfide is formed by $H_2S$ precipitation. The sulfide is then taken to the metal by hydrogen reduction.

## DEVELOPMENT AND PROCESS IMPROVEMENT

### Thermal Diffusion

A. E. Cameron
L. O. Love
W. R. Rathkamp
W. K. Prater
G. M. Banic, Jr.
J. W. Raleigh

The collection of gases in electromagnetic isotope separation equipment has received study both here in Oak Ridge and in several European installations. While small amounts of relatively pure isotopic material can be collected and recovered, it is not considered that this process can be applied to the repeated collection of relatively large amounts of product. The use of krypton or xenon – both comparatively rare and expensive gases – would require a recycling in the separator, which would be both inefficient and cumbersome. The demand for research quantities of separated isotopes of the rare gases and for appreciable amounts of $O^{18}$ and $O^{17}$ led to the decision to investigate thermal diffusion facilities to supplement the electromagnetic separators. The oxygen isotopes are of value in biological studies and probably also in corrosion investigations. The proposal that $Kr^{84}$ be accepted as the isotope to be used in the international standard of wavelength light source has placed that isotope high on the priority list.

The electromagnetic separation group has undertaken the construction of thermal diffusion columns for oxygen isotope production, or for the isotopes of other gases of which large quantities of starting material are available. The design of the installation is patterned to some extent on that which A. O. C. Nier had in operation at one time at the University of Minnesota. The plant there consisted of three parallel installations, each with nine columns 12 ft in length and graded in size in groups of three as a power economy measure. The hot central tubes in the larger columns were 6 cm in diameter, the intermediate ones were 3.34 cm, and the small ones were 2.22 cm. The diffusion gap was approximately 6 mm in each case. The total length of each cascade was 32 meters. The installation which is under construction here departs in some respects from these dimensions because of the design being controlled largely by availability of tubing in the desired diameters.

The length of the cascade is 55 meters. Diameters of the hot tubes are 9.5, 3.14, and 1.9 cm. Three each of the different sizes are connected in series. Stainless steel tubing with thin walls has been used throughout. Bellows provide for the rather considerable expansion of the hot tube relative to the cold wall. The hot central tube is maintained in an axial position by stainless steel setscrews, which were tapped into the cold wall and welded over after the central tube was properly positioned. The same kind of construction also positions the cold wall relative to the water jacket.

The different columns are connected in series with $^3/_{16}$-in. copper tubing to minimize dead volume. Gas will be caused to flow alternately backward and forward through this tubing by an adaptation of the *gasschaukel* introduced by Clusius.[3] The flow through the connecting tubing between columns was secured in Clusius' apparatus by alternately heating and cooling the reservoirs at the ends of the cascade. The cycle of this gas movement was 5 to 10 min. An arrangement of two bellows which are cam-driven by a slow-speed motor has been designed by W. R. Rathkamp. One bellows will be connected to each end of the cascade. The unwanted volume of the bellows has been filled with an aluminum slug, which eliminates the dead volume. Piping has been so arranged that columns can be isolated or sampling can be done between columns.

The operation of the columns will be determined, to a great extent, by the temperature gradient which can be maintained between the hot and cold walls. Preliminary measurements on experimentally heated tubes indicate that the desired 300°C

[3] K. Clusius, *Helv. Chim. Acta* 33, 2142 (1950).

gradient can be attained. Higher temperature would be very desirable. Hot platinum spirals are introduced into the connecting lines to permit establishment of equilibrium between the isotopic species in the gas.

The oxygen plant will be operated under non-equilibrium conditions, with effectively an infinite reservoir input to maintain the feed concentration constant. Draw-off may be continuous, or an inventory of enriched material may be accumulated at the product end. The precise method of operation remains to be determined when operating characteristics have been established. The krypton plant presents a different type of problem. Here the feed material is relatively rare and expensive, and the system must be operated with reservoirs at both ends and probably to essentially true equilibrium.

The plant for rare gases is designed to minimize internal volume. The cold wall is of 1.25-cm-ID copper tubing. A hot central Nichrome wire is used instead of the hot tube. The wire will be spaced at intervals by mica spiders, and direct current will be used for heating in order to avoid vibration induced by alternating-current heating. Eight columns are connected in series, giving a total length of 49 meters. As in the oxygen cascade, provision is made for isolating individual columns and for sampling between columns. A bellows pump will introduce a slow movement to remove gas from the dead volumes in the connecting tubing.

At the end of the period covered by this report, both plants were nearing completion, with only heaters and circulating pumps remaining to be installed prior to vacuum testing and operational tests.

### Inert Gas Collection in the Electromagnetic Separator

H. B. Greene

Previous work has shown that it is necessary to renew the surface of the metallic target in order to retain appreciable quantities of gas which arrive as energetic positive ions. Saturation of a metallic surface occurs with approximately 0.0025 ma·hr/cm$^2$. Various furnace arrangements were tried in previous experimentation in attempts to secure continuous and controllable evaporation of metal onto the cooled collecting surface. None of these methods of evaporation proved successful. Magnesium appeared to be a satisfactory metal, and an apparatus was designed which would permit long periods of operation, on the order of 100 hr, and allow evaporation at the rate of, say, 0.1 g/hr.

The type of collector used is shown in Fig. 17, with the beam-defining faceplate removed. A bellows is connected through a flexible line to the outside of the tank. Applying air pressure or vacuum permits the bellows to be expanded and contracted without interference from the magnetic field. The time cycle of the bellows operation is controlled by an electric clock mechanism and a three-way solenoid valve. The back and forth motion of the bellows actuates a ratchet mechanism which drives a knurled wheel. Magnesium ribbon is pressed against this knurled wheel by a spring-loaded idler wheel. Motion of the knurled wheel causes the magnesium ribbon to unwind from a coil and to project through a guide tube into a vaporization filament. The filament, a coil of 0.050-in.-thick tantalum wire, is wound on a $\frac{1}{4}$-in. diameter and consists of about five turns. A 50-amp d-c supply for the filament, with direction

**Fig. 17. Argon Receiver with Magnesium Vaporizer.**

of current and of winding of filament such that the magnetic field is not opposed, removes any difficulty of distortion. A 1-oz spool of ribbon, 0.008 × 0.125 in., can be loaded into the magazine.

A run of 98 hr was made with this apparatus. The magnesium evaporated onto a target of flattened, water-cooled, copper tubing. The $A^{36}$ beam was allowed to impinge upon this surface, while the $A^{40}$ impinged upon a water-cooled magnesium alloy plate which was tilted at a slight angle to the normal (0 deg) beam. Magnesium was sputtered from this plate and was collected upon the upper plate of copper, also water-cooled. The magnesium ribbon was fed for only 5 out of every 30 min. This was calculated to renew the surface sufficiently often for good collection.

A total of approximately 5 ma·hr of $A^{36}$ was monitored on this collector. The magnesium was scraped off the cold copper target and outgassed in high vacuum to release the contained gas. The gas was purified by passage over uranium metal chips at 800°C. This treatment removed all impurities except hydrogen, which was removed by exposing the gas to activated uranium that had been prepared by heating uranium hydride in high vacuum. Chemical purity of the recovered argon was approximately 99.5%. The portion of the target on which the most intense portion of the beam fell yielded 1.5 ml of gas of 70% $A^{36}$ and the other portion yielded 0.4 ml of 17% $A^{36}$. This corresponds to a recovery of 41% of the monitored beam. From the magnesium plate and from magnesium sputtered onto the cooled copper plate by the $A^{40}$ beam, approximately 100 ml of argon was recovered.

No further work on this method of collecting isotopes of inert gases is planned at the present time. Other methods of making such separations appear to be more attractive for production purposes. A completion report covering the work done on this project is in preparation.

# STABLE ISOTOPE CHEMISTRY

Boyd Weaver

## INTRODUCTION

Chemical refinement of 63 lots of isotopes of 12 elements was completed. Activities in the separation of rare earths included the preparation of yttrium earth concentrate from gadolinite, the determination of distribution coefficients for gadolinium and yttrium between tributyl phosphate and nitric acid of various concentrations, and the relative distribution of various rare earths in multistage contactors under various conditions.

## CHEMICAL REFINEMENT OF ISOTOPES

W. C. Davis F. B. Thomas
R. L. Bailey

Progress was made in the chemical refinement of isotopes of 16 elements: Mg, S, Ti, Cu, Zn, Ge, Br, Rb, Sr, Mo, Ag, Cd, Ba, Sm, Eu, and W. The following discussion concerns only those elements of which the current series have been completed.

### Magnesium

Magnesium metal, alloyed with copper, was dissolved from the copper pockets with dilute acid, giving solutions containing many times as much copper as magnesium. Purification was accomplished by the following general procedure, with some modifications to fit special cases.

1. Evaporate the solutions to crystals of $Cu(NO_3)_2 \cdot 6H_2O$ to remove excess $HNO_3$.
2. Redissolve in $H_2O$, add $H_2SO_4$, and electrolyze to remove most of the copper. In some cases a part of the copper has been removed by crystallization as $CuSO_4$, without significant loss of magnesium.
3. Evaporate solutions to fuming $H_2SO_4$ to remove all $HNO_3$.
4. Dissolve in $H_2O$ and make $H_2S$ precipitations in both acid and ammoniacal conditions.
5. Remove ammonium salts by boiling with aqua regia.
6. Evaporate to fumes of $H_2SO_4$.
7. Dissolve sulfates in $H_2O$ and precipitate magnesium with 8-hydroxyquinoline.
8. Filter and wash to remove all sulfate.
9. Dry precipitate, cover with oxalic acid, and ignite to MgO by gradual heating to 1000°C.
10. Dissolve the still impure MgO in dilute $H_2SO_4$, filter, digest with 90% methanol, and filter off $CaSO_4$.
11. Evaporate filtrate to dryness, dissolve in $H_2O$, and repeat $H_2S$ precipitations.
12. Reprecipitate magnesium oxyquinolate and ignite to MgO.

The largest lot of $Mg^{24}$ was not precipitated as the oxyquinolate because of the difficulty of removing sulfate. The dry sulfate crystals were dissolved by long extraction with about 90% methanol and filtered from the $CaSO_4$. The solution was evaporated to dryness and the $MgSO_4$ crystals were ignited to 850°C. Production data on the series are given in Table 10.

### Bromine

Bromine isotopes were collected on magnesium turnings in copper receivers. The following procedure has been established for recovery and purification.

1. Dissolve most of the $MgBr_2$ with hot $H_2O$.
2. Complete recovery by leaching with cold dilute $HNO_3$.
3. Add $AgNO_3$ solution in excess and combine the precipitates on a fritted glass filter.
4. Dissolve AgBr through the filter with $NH_4OH$.
5. Reprecipitate AgBr by adding $HNO_3$ until fairly acid.
6. Filter and redissolve with excess $NH_4OH$.
7. Pour solution through a column of Dowex 50 cation-exchange resin in the ammonium state and wash with $H_2O$. Silver is left on the resin.
8. Evaporate the solution to dryness slowly in a plastic beaker, transfer the solid $NH_4Br$ to a porcelain dish, and dry overnight at 200°C.

Production data on this series are given in Table 11.

### Strontium

Strontium isotopes were collected in water-cooled copper receivers. Most of the strontium spattered from the surface of initial contact to the opposite surface of the receiver, along with a large quantity of copper. The strontium was recovered quantitatively only by dissolving the resulting alloy and part of the receiver with $HNO_3$. The $Sr^{84}$ was first purified by a combination of electrolysis, sulfide precipitations, and conversion to the carbonate. This left the common impurities Ca, Ba, Mg, and Pb and traces of other elements. Crystallization of $Ba(NO_3)_2$ from 80% $HNO_3$ still left barium and lead. The lead was removed by anode deposition

TABLE 10. PRODUCTION DATA ON MAGNESIUM ISOTOPES

| Isotope | Lot No. | Weight of MgO (g) | Impurities Found (%) | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Al | Ca | Cr | Cu | Fe | K | Mo | Na | Ni | Pb | Si |
| 24 | HM 928(a) | 262.00* | | 0.1 | | | | 0.01 | | 0.02 | | | |
| 24 | HM 928(b) | 11.2234 | <0.05 | 0.05 | | 0.1 | <0.02 | | | 0.01 | | | <0.05 |
| 24 | HM 928(c) | 11.1470 | | 0.05 | | | <0.02 | | | | | | |
| 25 | HM 929(a) | 9.2953 | <0.05 | 0.1 | | 0.01 | 0.02 | | | | | | |
| 25 | HM 929(b) | 1.0327 | <0.01 | 0.01 | | 0.02 | | 0.01 | | 0.02 | | 0.01 | <0.01 |
| 25 | HM 929(c) | 1.1873 | | 0.1 | | 0.0025 | 0.005 | | | | | | |
| 26 | HM 930(a) | 10.3989 | | 0.1 | | 0.04 | <0.02 | | | | 0.1 | | |
| 26 | HM 930(b) | 0.7780 | | 0.08 | <0.01 | | <0.005 | <0.004 | <0.01 | | | | <0.01 |
| 26 | HM 930(c) | 1.5329 | <0.01 | 0.07 | | <0.0025 | <0.005 | 0.004 | | 0.02 | | | <0.01 |

*$MgSO_4$.

TABLE 11. PRODUCTION DATA ON BROMINE ISOTOPES

| Isotope | Lot No. | Weight of $NH_4Br$ (g) | Impurities Found (%) | | | | |
|---|---|---|---|---|---|---|---|
| | | | Cu | K | Si | Mg | Na |
| 79 | HG 896(a) | 4.164 | <0.02 | 0.01 | <0.05 | | |
| 81 | HG 897(a) | 4.186 | <0.02 | 0.01 | <0.05 | <0.02 | 0.01 |

and the barium by chromate precipitation; final purification was attained by precipitating $Sr(NO_3)_2$ twice from 80% $HNO_3$. Based on this experience the following simplified procedure was applied to the other isotopes.

1. Remove copper and lead by electrolysis, with care that deposition of lead is complete.
2. Reduce volume and acidity by evaporation.
3. Buffer the solution with ammonium acetate.
4. Add $(NH_4)_2CrO_4$ in slight excess and filter off $BaCrO_4$.
5. Reduce to a minimum volume and add 95% fuming $HNO_3$ slowly with rapid agitation until the $HNO_3$ content reaches 80%. Let settle for 30 min.
6. Filter on fritted glass, dissolve through the filter with hot $H_2O$, transfer to a beaker, and evaporate to a low volume.
7. Repeat the crystallization from 80% $HNO_3$.
8. Dry the crystals on a steam bath and then in an oven at 130 to 140°C.

Production data are given in Table 12. Material recovered from the separator faceplates and designated as (b) was not given the final nitrate treatment.

### Molybdenum

Molybdenum isotopes were collected in copper receivers and were present as deposits which could be removed only by dissolving away much of the supporting copper with $HNO_3$. Much of the molybdenum settled as a black sludge. Refinement was accomplished by the following procedure.

1. Evaporate solution to near dryness on a steam bath, thus dissolving the molybdenum and reprecipitating most of it as $MoO_3$.
2. Dilute with $H_2O$ and filter.
3. Dissolve precipitate and paper in hot concentrated $H_2SO_4$ and $HNO_3$, dilute, and filter. Reserve solution.
4. Dilute filtrate from (2), cool to 10°C, and precipitate molybdenum by adding a cold solution of 2% $\alpha$-benzoinoxime in ethanol. Ensure complete oxidation of molybdenum by addition of bromine water.

TABLE 12. PRODUCTION DATA ON STRONTIUM ISOTOPES

| Isotope | Lot No. | Weight of $Sr(NO_3)_2$ (g) | Impurities Found (%) | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Al | Ba | Ca | K | Li | Mg | Mo | Na | Pb | Si |
| 84 | HG 898(a) | 4.989 | | 0.1 | | | | <0.02 | | <0.01 | <0.1 | <0.05 |
| 86 | HG 899(a) | 7.038 | <0.05 | 0.05 | | | | <0.02 | <0.02 | | <0.1 | 0.05 |
| 86 | HG 899(b) | 1.111 | 0.05 | 0.05 | <0.1 | 0.01 | | 0.02 | | <0.01 | | |
| 87 | HG 900(a) | 7.822 | | 0.1 | | | | <0.02 | | | | <0.05 |
| 87 | HG 900(b) | 1.600 | <0.05 | 0.1 | | 0.03 | | <0.02 | | 0.01 | 0.2 | |
| 88 | HG 901(a) | 36.102 | | | | | <0.01 | <0.02 | | 0.01 | | |
| 88 | HG 901(b) | 11.522 | <0.05 | 0.1 | | 0.05 | | <0.02 | | 0.01 | | |

5. After 15 to 30 min of standing, filter precipitate on medium fritted filter covered by a thin layer of macerated filter paper and wash with cold 1% $H_2SO_4$ containing $\alpha$-benzoinoxime.

6. Carefully ignite precipitate at 525 to 550°C, dissolve in concentrated $H_2SO_4$, dilute, and filter.

7. Combine with solution from (3), dilute to 2 liters, add 100 ml of 6% $H_2SO_3$ to reduce chromium and vanadium, and boil off excess.

8. Cool to 10°C and precipitate molybdenum with $\alpha$-benzoinoxime.

9. Filter, ignite, redissolve in hot $H_2SO_4$, dilute, filter, reprecipitate.

10. Repeat (9).

11. Ignite carefully at 525 to 550°C in a Vycor crucible to $MoO_3$.

Production data for the series are given in Table 13.

### Silver

The silver isotopes were collected partly on copper receivers and partly on graphite inserts within the copper receivers. Most of the latter material was spattered from the graphite to the copper, where it adhered tightly as a silver-carbon compound insoluble in $HNO_3$. It was recovered by ignition with a torch and by dissolving away the copper base and igniting as a nitrate solution. The silver was then purified by the following procedure.

1. Precipitate AgCl by adding a slight excess of HCl to the nitrate solution. Filter on fritted glass and wash with dilute $HNO_3$.

2. Dissolve AgCl through the filter with $NH_4OH$.

3. Reprecipitate AgCl by adding an excess of $HNO_3$ and a little HCl.

4. Filter and redissolve with $NH_4OH$.

5. Add an excess of hydroquinone, stir for 5 min, and centrifuge the precipitated silver.

6. Wash several times with $H_2O$ and then with ethanol and acetone to remove all organic matter.

7. Dry overnight at 110°C in the centrifuge bottle, transfer to a porcelain dish, and heat at 300°C for about 1 hr. Avoid sintering.

Production data are given in Table 14.

### Samarium

Samarium isotopes were collected in standard graphite receivers. The impure oxide obtained by ignition was purified by the standard procedure which has been developed for rare earths.

1. Dissolve solids in $HNO_3$.

2. Repeatedly precipitate with $NH_4OH$ and redissolve in acid until all copper blue color has disappeared.

3. Dissolve hydroxide in citric acid, dilute, and make sulfide precipitations from both acid and ammoniacal solutions. Filter.

4. Precipitate $Sm_2(C_2O_4)_3$. Filter on paper and ignite.

5. Dissolve $Sm_2O_3$ in $HNO_3$, dilute to approximately 0.5 *N* $HNO_3$, and precipitate $Sm_2(C_2O_4)_3$ with $H_2C_2O_4$.

6. Filter oxalate on paper and ignite to $Sm_2O_3$ in porcelain at 900°C.

Since spectrographic analysis showed the presence of an undesirable amount of neodymium in the $Sm^{144}$, it was further purified by elution

from a cation exchange column with 5% citric acid at pH 2.9. A total of 510 mg of $Sm_2O_3$ free of neodymium was obtained, with all neodymium being concentrated into an additional 65 mg of oxide.

Production data for the series are given in Table 15.

## PROTACTINIUM CHEMISTRY

F. M. Scheitlin Boyd Weaver

Progress in the chemistry of protactinium has been reported in the quarterly reports of the Homogeneous Reactor Project.

TABLE 13. PRODUCTION DATA ON MOLYBDENUM ISOTOPES

| Isotope | Lot No. | Weight of $MoO_3$ (g) | Impurities Found (%) | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | Ag | Al | Ca | Fe | Mg | Ni | Pb | Si | Sn | Ti |
| 92 | HF 889(a) | 6.527 | Trace | | <0.05 | <0.02 | <0.02 | | <0.1 | 0.2 | | 0.02 |
| 94 | HF 890(a) | 5.122 | | | | | | | | | | <0.02 |
| 95 | HF 891(a) | 2.426 | Trace | 0.05 | <0.01 | 0.02 | <0.02 | | | 0.05 | | <0.02 |
| 95 | HF 891(b) | 2.281 | Trace | 0.05 | 0.01 | 0.02 | <0.02 | | | <0.05 | | <0.02 |
| 95 | HF 891(c) | 2.331 | Trace | 0.05 | <0.01 | 0.2 | <0.02 | | | <0.05 | | <0.02 |
| 95 | HF 891(d) | 2.057 | Trace | 0.05 | 0.01 | 0.02 | <0.02 | | | <0.05 | | <0.02 |
| 96 | HF 892(d) | 10.469 | | | <0.02 | 0.02 | <0.02 | | <0.1 | 0.1 | 0.3 | |
| 97 | HF 893(a) | 2.305 | | | <0.01 | | 0.02 | | | | | <0.02 |
| 97 | HF 893(b) | 1.493 | | | <0.02 | | 0.02 | | <0.1 | 0.1 | | 0.02 |
| 97 | HF 893(c) | 1.901 | | | 0.01 | | | | | <0.05 | | <0.02 |
| 97 | HF 893(d) | 1.672 | | | | | | | | <0.05 | | <0.02 |
| 98 | HF 894(a) | 15.108 | | | 0.01 | | <0.02 | | | <0.05 | | |
| 100 | HF 895(a) | 1.854 | | | | | <0.01 | | | | | 0.01 |
| 100 | HF 895(b) | 1.739 | | 0.05 | <0.05 | 0.02 | <0.02 | | | <0.05 | | |
| 100 | HF 885(c) | 1.148 | | | | | <0.01 | 0.05 | | 0.05 | | 0.02 |
| 100 | HF 895(d) | 0.941 | | | | | | | | <0.02 | | 0.02 |

TABLE 14. PRODUCTION DATA ON SILVER ISOTOPES

| Isotope | Lot No. | Weight of Ag (g) | Impurities Found (%) | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | | Al | B | Cu | Fe | Mg | Pb | Si |
| 107 | HO 936(a) | 24.443 | | | <0.05 | | <0.02 | | <0.05 |
| 107 | HO 936(b) | 23.327 | 0.1 | | | <0.02 | 0.03 | <0.1 | 0.2 |
| 107 | HO 936(c) | 6.282 | | <0.01 | <0.05 | | <0.02 | | 0.1 |
| 109 | HO 937(a) | 21.370 | | | | | | | <0.05 |
| 109 | HO 937(b) | 19.308 | | | No impurities detected | | | | |
| 109 | HO 937(c) | 5.701 | | | | | <0.05 | | <0.05 |

TABLE 15. PRODUCTION DATA ON SAMARIUM ISOTOPES

| Isotope | Lot No. | Weight of $Sm_2O_3$ (g) | Impurities Found (%) | | | | |
|---|---|---|---|---|---|---|---|
| | | | Ca | Eu | Fe | Mg | Nd |
| 144 | HI 907(a) | 0.510 | 0.05 | <0.05 | 0.02 | | |
| 147 | HI 908(a) | 3.004 | 0.03 | <0.05 | | | 0.05 |
| 148 | HI 909(a) | 2.856 | 0.03 | <0.05 | | | 0.1 |
| 149 | HI 910(a) | 3.560 | | | | <0.02 | |
| 150 | HI 911(a) | 2.306 | 0.03 | 0.2 | | | 0.2 |
| 152 | HI 912(a) | 8.540 | 0.03 | 0.1 | | | |
| 154 | HI 913(a) | 6.053 | No impurities detected | | | | |

## RARE-EARTH CHEMISTRY

F. A. Kappelmann L. J. Royer
Boyd Weaver

### Raw Material Processing

Most of the supply of gadolinite ore on hand has been leached with nitric acid. By countercurrent extraction with tributyl phosphate the yttrium-earth content has been separated from iron, beryllium, and most of the cerium earths.

### Separations by Liquid-Liquid Extraction

In order to obtain basic information on the behavior of rare earths and associated elements, distribution coefficients for various rare earths between tributyl phosphate and nitric acid are being determined under various conditions. Since yttrium is a major constituent of all yttrium-earth concentrates and gadolinium is the most readily available of the heavier rare earths, the first work is being done with these two elements. Typical data from equal-volume separatory-funnel shakeouts are shown as concentration isotherms in Fig. 18. It is observed that, at high acidity (12.3 *N*), yttrium extracts preferentially to gadolinium and that the isotherms are not rectilinear at high concentrations. With an $HNO_3$ concentration of 4 *N*, the relative extractabilities of gadolinium and yttrium are reversed and the isotherms are straighter.

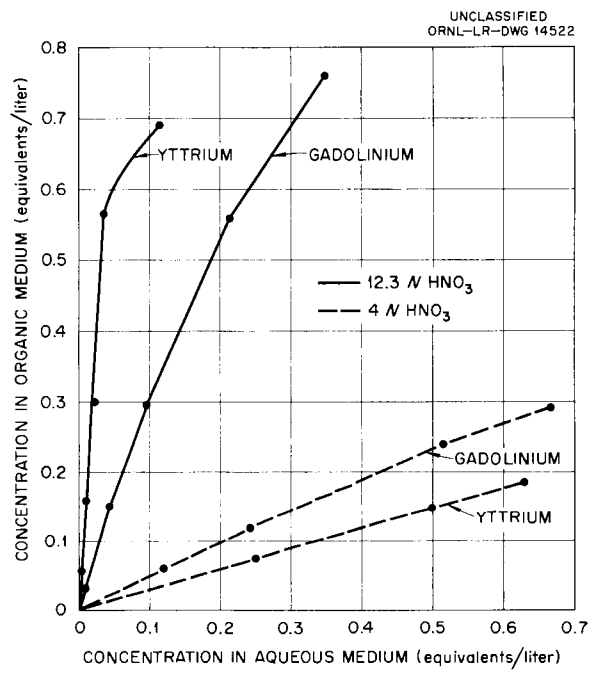



**Fig. 18. Distribution of Gadolinium and Yttrium Between TBP and $HNO_3$.**

### Process Development

Neodymium, samarium, and gadolinium of high quality have been separated by a countercurrent system in which the aqueous stripping feed and organic streams were of the same order of magnitude. The increase in deviation of distribution isotherms from linearity with increase in concentration and atomic number reduces the effectiveness of this process to near impracticability for purification of the heavier elements. The great preponderance of yttrium in heavy-earth concentrates makes its prior removal essential, especially if the process for separation of the other elements requires low concentrations. The very high dis-

tribution coefficients of the heavy elements between pure TBP and very strong $HNO_3$ result in unwieldy flow ratios. The lack of external reflux inherently limits the degree of separation attainable, though it allows maximum throughput. In consideration of these facts, an extraction column pilot plant and an 11-stage mixer settler are being operated under various modifications of the basic process. The major changes applied or projected include dilution of the TBP, operation with very small flow rate of highly concentrated feed, a double plant in which yttrium is shifted to different positions relative to the lanthanons (lanthanide rare earths), operation at low concentrations, operation with the TBP so highly saturated with rare earths that it behaves as an exchange medium, and application of external reflux.

Typical behavior with the conventional open-end process is illustrated by Experiment I, in which gadolinite concentrate was passed through the pulse column pilot plant. Operating conditions and results are given in Table 16. The aim was to produce a concentrate of ytterbium and the neighboring elements separated from a concentrate of dysprosium. The spectrographic product analyses given are of spot samples taken when the system was believed to be in equilibrium. It is obvious that, while there was separation in the desired direction, it was not nearly so selective as desirable.

In Experiment II the feed was a composite from the organic effluent of Experiment I. It was concentrated to 300 g of oxide per liter in 1.7 *N* $HNO_3$ and fed at a rate less than 0.01 of the total aqueous flow. Operating conditions and results are given in Table 17. While the organic effluent was considerably enriched in ytterbium, the major part of this element was still in the aqueous effluent. In a current experiment the position of feed introduction has been altered.

Experiment III was performed in the mixer-settler with 11 mechanical stages, with the acidity 10.4 *N* $HNO_3$. The feed and flow ratios were the same as for Experiment II. Operating conditions and results are presented in Table 18. The mixer-settler appears to have been more effective than the pulse column, with many more possible stages.

In Experiment IV the acidity was lowered to 4 *N* and the flow ratios were adjusted to correct for the decrease in distribution coefficients. Operating conditions and analyses are given in Table 19.

**TABLE 16. PULSE COLUMN EXTRACTION OF GADOLINITE CONCENTRATE WITH DIVIDED AQUEOUS FLOW – EXPERIMENT I**

Aqueous medium: 10.4 *N* $HNO_3$; flow rate, 2.8 liters/hr

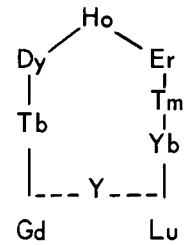
Feed: 25 g of oxide per liter; 15 *N* $HNO_3$; flow rate, 5.7 liters/hr

Organic medium: TBP equilibrated with 10.4 *N* $HNO_3$; flow rate, 4.0 liters/hr

| Rare-Earth Oxide | Analysis (%) | | |
|---|---|---|---|
| | Aqueous Effluent | Feed | Organic Effluent |
| $Nd_2O_3$ | 0.7 | 2 | |
| $Sm_2O_3$ | 1.5 | 5 | |
| $Eu_2O_3$ | Trace | Trace | |
| $Gd_2O_3$ | 3.6 | 1.2 | 0.5 |
| $Tb_4O_7$ | 1.5 | 1 | Trace |
| $Dy_2O_3$ | 10.8 | 5 | 1.6 |
| $Ho_2O_3$ | 2.5 | 1.5 | Trace |
| $Er_2O_3$ | 6.3 | 5 | 3.1 |
| $Tm_2O_3$ | | 0.9 | 0.25 |
| $Yb_2O_3$ | Trace | 4.5 | 5 |
| $Lu_2O_3$ | | 0.8 | 1.2 |
| $Y_2O_3$ | 74* | 73* | 88* |
| Fraction of total | 0.41 | | 0.59 |

*By difference.

As an interesting observation, not only was the extraction of yttrium very similar to that of gadolinium but also the order of extraction of the heavier lanthanons was altered. The order appears to be that represented below:




Another experiment in which the feed flow rate was increased from 25 to 80 ml/hr gave essentially

**TABLE 17. PULSE COLUMN EXTRACTION WITH CONSTANT AQUEOUS FLOW – EXPERIMENT II**

Aqueous medium: 10.4 *N* $HNO_3$; flow rate, 18 liters/hr

Feed: 300 g of oxide per liter; 1.7 *N* $HNO_3$; flow rate, 0.15 liter/hr

Organic medium: TBP equilibrated with 10.4 *N* $HNO_3$; flow rate, 3 liters/hr

| Rare-Earth Oxide | Analysis (%) | | |
|---|---|---|---|
| | Aqueous Effluent | Feed | Organic Effluent |
| $Gd_2O_3$ | Trace | 0.3 | |
| $Dy_2O_3$ | 1.7 | 1.1 | 0.5 |
| $Ho_2O_3$ | 0.7 | 0.6 | Trace |
| $Er_2O_3$ | 4 | 3.3 | 2.7 |
| $Tm_2O_3$ | 1.1 | 1.3 | 1.0 |
| $Yb_2O_3$ | 7.1 | 7.5 | 23 |
| $Lu_2O_3$ | 0.5 | 1.4 | 5.0 |
| $Y_2O_3$ | 84* | 84* | 66* |
| Fraction of total | 0.83 | | 0.17 |

*By difference.

**TABLE 18. MIXER-SETTLER EXTRACTION – EXPERIMENT III**

Aqueous medium: 10.4 *N* $HNO_3$; flow rate, 3.00 liters/hr

Feed: 330 g of oxide per liter; 0.17 *N* $HNO_3$; flow rate, 0.025 liter/hr

Organic medium: TBP equilibrated with 10.4 *N* $HNO_3$; flow rate, 0.500 liter/hr

| Rare-Earth Oxide | Analysis (%) | | |
|---|---|---|---|
| | Aqueous Effluent | Feed | Organic Effluent |
| $Gd_2O_3$ | Trace | 0.3 | |
| $Tb_4O_7$ | | Trace | |
| $Dy_2O_3$ | 0.9 | 1.1 | <0.5 |
| $Ho_2O_3$ | 0.7 | 0.6 | <0.5 |
| $Er_2O_3$ | 3.2 | 3.3 | 2.6 |
| $Tm_2O_3$ | 0.7 | 1.3 | 2.2 |
| $Yb_2O_3$ | 3.1 | 7.5 | 33.5 |
| $Lu_2O_3$ | Trace | 1.4 | 4.7 |
| $Y_2O_3$ | 91* | 84* | 56* |
| Fraction of total | 0.74 | | 0.26 |

*By difference.

the same results. This apparently eliminates one method of separating yttrium widely from the heavier lanthanons.

In the use of diluents to decrease distribution coefficients the difficulty was encountered that at high acidities, where dilution is needed, three liquid phases were formed. There are hydrocarbon diluents, however, sufficiently stable to be used with very strong nitric acid.

Mixer-settler experiments with external reflux have not been carried to equilibrium. Extensive development and modification of evaporators and pumping systems have been required to obtain the necessary precise control of process streams.

Development of the extraction process has been greatly aided by consultation on engineering problems with J. M. Googin of the Y-12 staff.

### Analytical Development

Control of any process involving solutions of rare earths requires frequent measurements of both concentrations and compositions of process streams. The usual technique involves the time-consuming process of conversion to oxide followed by some physical analytical method such as spectrophotometry or spectrography. A new method is now being developed for measuring concentrations in equivalency units by simply titrating solutions with a standard alkali solution. End points may be observed by the use of a pH meter or suitable indicators. Only preliminary work has been done with solutions of nitrates, chlorides, sulfates, and perchlorates. Figure 19 shows the results of titrating 0.1060 g of $Gd_2O_3$ with 0.1000 *N* NaOH in a 200-ml volume. While there is no completely flat plateau at either end of the titration, the results are sufficiently accurate for most purposes. The use of methyl red and thymol blue indicators instead of a pH meter gives only slightly higher results. Titration of chlorides has been equally accurate. Titration of sulfates and perchlorates must be performed very slowly in order to maintain equilibrium, and the end points are very indefinite. There is no definite color change with thymol blue.

**TABLE 19. MIXER-SETTLER EXTRACTION – EXPERIMENT IV**

Aqueous medium: 4 *N* $HNO_3$; flow rate, 0.900 liter/hr

Feed: 330 g of oxide per liter; 0.17 *N* $HNO_3$; flow rate, 0.025 liter/hr

Organic medium: TBP equilibrated with 4 *N* $HNO_3$; flow rate, 1.80 liters/hr

| Rare-Earth Oxide | Analysis (%) | | |
|---|---|---|---|
| | Aqueous Effluent | Feed | Organic Effluent |
| $Gd_2O_3$ | 7.5 | 0.3 | 6.0 |
| $Tb_4O_7$ | Trace | Trace | 2.8 |
| $Dy_2O_3$ | Trace | 1.1 | 1.4 |
| $Ho_2O_3$ | | 0.6 | 0.6 |
| $Er_2O_3$ | 1.1 | 3.3 | 3.1 |
| $Tm_2O_3$ | 0.7 | 1.3 | 1.2 |
| $Yb_2O_3$ | 7.6 | 7.5 | 10.4 |
| $Lu_2O_3$ | 1.3 | 1.4 | 1.2 |
| $Y_2O_3$ | 81* | 84* | 73* |
| Fraction of total | 0.20 | | 0.80 |

*By difference.

Figure 20 gives the results obtained by titration of 1.000 g of $Gd_2O_3$ as nitrate with 1.000 *N* NaOH in 200 ml. Some care was necessary to avoid pre-precipitation of hydroxide in neutralizing the excess acid, but the titration gave accurate results.

An interesting finding was that the volumes of alkali measured are exactly 0.80 times the stoichiometrical amounts for the theoretical conversion of nitrate to hydroxide. It is evident that the precipitate is a basic nitrate with the formula $4Gd(OH)_3 \cdot Gd(NO_3)_3$. Equivalent results were obtained by titration of yttrium salts.

One possible application of the titration is the determination of average atomic weights of mixtures. This has been done in the past by weighing the mixtures in two chemical forms or by weighing an oxide and titrating the oxalate. Titration makes it possible to make measurements in a much shorter time. This is especially useful with mixtures of yttrium and the heavy earths.

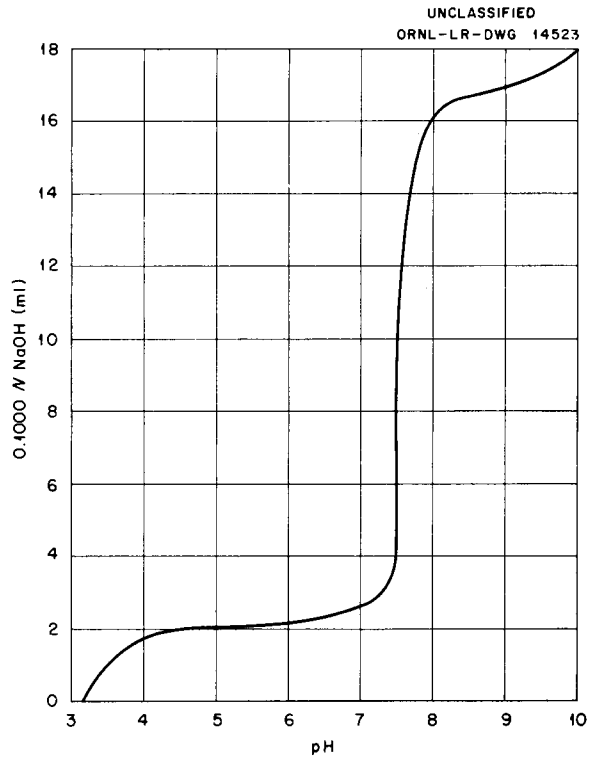



**Fig. 19. Titration of $Gd_2O_3$ with 0.1000 *N* NaOH.**

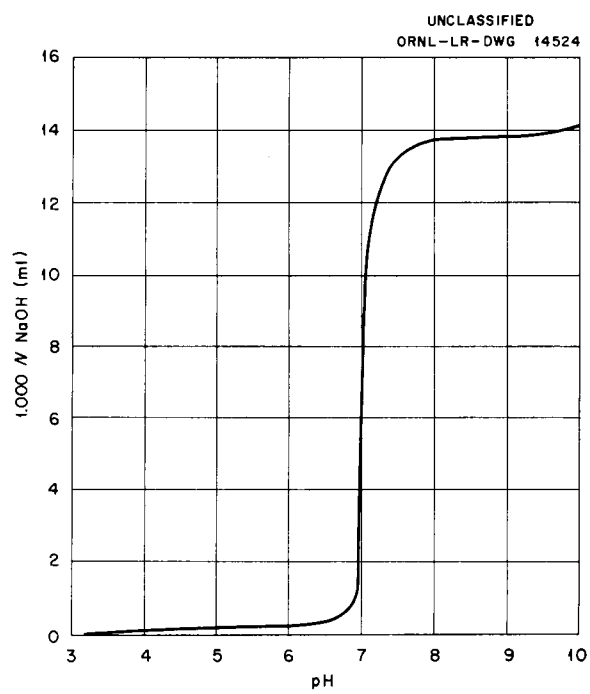



**Fig. 20. Titration of $Gd_2O_3$ with 1.000 *N* NaOH.**

# UTILIZATION OF ENRICHED STABLE ISOTOPES

P. S. Baker F. R. Duncan

## TARGET PREPARATION AND SPECIAL SERVICES

The requests for assistance by research laboratories which use stable isotopes continue to increase. The needs during the period covered by this report made it necessary not only to develop new procedures but also to re-employ procedures such as those previously discussed.[1]

### Target Preparation

In order to supply isotopes for elemental targets of tungsten, germanium, zinc, titanium, and chromium for neutron-scattering experiments at Westinghouse Research Laboratories, it became necessary in each case to develop methods for reducing the inventory material to the metallic state. The usual care was taken to prevent undue loss of the valuable isotopic samples. These procedures are described briefly:

1. *Tungsten.* – Tungsten trioxide can be reduced satisfactorily in a hydrogen furnace by heating at 950°C for 3 hr. The metal powder is then compacted at 45 tsi and sintered to give suitable $1 \times \frac{1}{3}$ in. coupons (thickness determined by the amount of sample). Twelve individual coupons of $W^{182}$, $W^{183}$, $W^{184}$, and $W^{186}$, weighing from 1 to 3 g each, were prepared in this manner.

2. *Germanium.* – The reduction of $GeO_2$ can also be carried out in a hydrogen furnace at elevated temperature, with compaction and sintering similar to that for tungsten. Fifteen individual coupons of the five germanium isotopes, weighing from 1 to 6 g each, were prepared in this way.

3. *Zinc.* – The preparation of metallic zinc targets involved reduction of ZnO with tantalum powder at 950°C for 4 hr, with the zinc being distilled off as it was formed. It was then collected and compressed into $1 \times \frac{1}{3}$ in. coupons with a pressure of 30 tsi. Elemental $Zn^{66}$ and $Zn^{68}$ were prepared in this way, the coupons weighing 0.9 and 1.6 g, respectively.

4. *Titanium.* – Metallic titanium was produced by the thermochemical reduction of $TiO_2$ with elemental calcium in an inert atmosphere at 800°C. The excess unreacted calcium and the calcium oxide formed during the reaction were leached out with dilute acid and the resulting titanium powder was filtered off, dried, and compacted at 25 tsi. Even with considerable leaching it was impossible to reduce the calcium content of the final titanium below about 2%. However, the procedure is much simpler than is the reduction of $TiCl_4$ with magnesium, and, since the resulting titanium was acceptable to the user, this method was used. Twenty-one individual coupons of the five isotopes of titanium were prepared, with weights ranging from 0.5 to 12 g.

5. *Chromium.* – The preparation of $Cr^{52}$ targets was almost identical with that for titanium described above. Three coupons of 9 g each were prepared by this method.

### Special Procedures

A number of other special conversions were made, three of which are listed below:

1. *Potassium metal.* – A 2-g sample of $K^{39}$ metal was prepared by thermochemical reduction of $K^{39}Cl$ with elemental calcium at 700°C. The procedure is similar to the technique mentioned earlier for the preparation of metallic rubidium.[2]

2. *Cadmium foil.* – A 2-cm square foil of $Cd^{111}$ metal was prepared by reduction of CdO with carbon at about 850°C. The cadmium distilled off and after agglomeration was rolled into a sheet approximately 0.0015 in. thick.

3. *Lithium-7 metal.* – Pile oscillator targets were prepared from barium-reduced,[3] distilled[4] lithium metal, diluted with normal lithium metal to give $Li^7$ concentrations of 99.8, 99.6, and 99.4%.

In addition to these new procedures for preparations, there were a number of targets and conversions made for elements such as iron, nickel, copper, and lithium, the techniques for which have been reported previously.

---

[1] P. S. Baker *et al.*, *Stable Isotope Research and Production Semiann. Prog. Reps. Nov. 20, 1954, and May 20, 1955*, ORNL-1829 and ORNL-1908, p 42.

[2] P. S. Baker *et al.*, *Stable Isotope Research and Production Semiann. Prog. Rep. Nov. 20, 1954*, ORNL-1829, p 45.

[3] P. S. Baker, F. R. Duncan, and H. B. Greene, *Science* 119, 469 (1954).

[4] P. S. Baker, F. R. Duncan, and H. B. Greene, *Science* 118, 778 (1953).

### Cooperative Calutron-Cyclotron Program

The 86-in. cyclotron was shut down for most of the period covered by this report; therefore no new runs were made. However, the technique for coating cyclotron targets with magnesium has been improved,[5] and it is now possible to obtain thicknesses of from 0.010 to 0.012 in. over an area 3 × 4 in. This improved method was actually utilized to make eight targets of various thicknesses and covering various areas with the three isotopes of magnesium for use by the 63-Inch Cyclotron Group. It is planned that the production-type target for converting $Ca^{48}$ to $Ca^{47}$ (4.8-day) be prepared, bombarded, and processed soon.

## ISOTOPE DISTRIBUTION

P. S. Baker

A significant change in the isotope distribution program was effected during the period covered by this report in that the Atomic Energy Commission approved the domestic sales of electromagnetically enriched stable isotopes. Allocation by the Commission is no longer necessary, and arrangements for purchase or loans are made directly with the Stable Isotopes Division. Coincident with the change in procedure was the transfer of the distribution and accountability of enriched $B^{10}$ from the Radioisotopes Sales Department to this Division.

The new procedures worked out by the Commission encourage purchase of isotopes by the users wherever possible but at the same time permit loans of those isotopes specified by the AEC as "rare and expensive" provided that they will not be diluted or contaminated by the user. Listed below are those isotopes which are not considered as either rare or expensive and which, for that reason, are for sale only:

| | |
|---|---|
| $Ba^{138}$ | All enrichments |
| $B^{10}$ | All enrichments |
| $B^{11}$ | Any enrichment less than 90% |
| $Cd^{113}$ | Any enrichment less than 26% |
| $Cd^{116}$ | Any enrichment less than 25% |
| $Ca^{40}$ | Any enrichment less than 99% |
| $Cr^{52}$ | Any enrichment less than 97% |
| $Hf^{174}$ | Any enrichment less than 0.5% |
| $Hf^{176}$ | Any enrichment less than 11% |
| $Hf^{178}$ | Any enrichment less than 35% |
| $Hf^{180}$ | Any enrichment less than 50% |
| $Fe^{56}$ | All enrichments |
| $Pb^{204}$ | Any enrichment less than 0.6% |
| $Li^{6}$ | All enrichments |
| $Nd^{142}$ | Any enrichment less than 57% |
| $Ni^{58}$ | All enrichments |
| $Sn^{116}$ | Any enrichment less than 60% |
| $Sn^{118}$ | Any enrichment less than 45% |
| $Sn^{120}$ | All enrichments |
| $Ti^{48}$ | Any enrichment less than 98% |

During the three-month transition period, in which no loans of isotopes were made, the number of shipments was comparable to that of a year ago. During December 1955 the 3000th shipment of electromagnetically enriched isotopes was distributed from the Division, and the total through May 20, 1956, was 3232 shipments. It is of interest to note that the number of shipments (75) for the first three weeks of May 1956 was greater than that for any single month in the ten-year history of the Division. Experience thus far under the new sales policy for stable isotopes indicates, in general, an increase in shipments, both domestic and foreign, an increase in reimbursement toward production and handling, and a favorable reception by stable isotope users to the purchase of enriched isotopes. Comments with reference to shipments do not include an appreciable number of shipments of boron and lithium.

Table 20 indicates the distribution of shipments as well as the number of shipments by months for the six-month period ending May 20, 1956. It will be noted that targets and special services have been provided for about 12% of the total. The number of foreign shipments for this period is 30% greater than the total of all foreign shipments during the previous 15 months.

It is perhaps significant that the number of off-project users continues to increase relative to on-project users. Table 21 shows the trend during the ten years of distribution of enriched isotopes.

Table 22 is a detailed list of the isotope shipments during the period from November 20, 1955, through May 20, 1956.

[5] P. S. Baker and F. R. Duncan, *Stable Isotopes Research and Production Semiann. Prog. Rep. Nov. 20, 1955*, ORNL-2028, p 44.

TABLE 20. TOTAL SHIPMENTS OF ISOTOPES – NOVEMBER 20, 1955, THROUGH MAY 20, 1956

| Month | On-Project | Off-Project | Foreign | Total | Targets and Special Services |
|---|---|---|---|---|---|
| November | 2 | 3 | | 5 | 1 |
| December | 11 | 29 | 13 | 53 | 4 |
| January | 17 | 18 | 1 | 36 | 7 |
| February | 10 | 14 | 8 | 32 | 2 |
| March | 17 | 17 | 10 | 44 | 4 |
| April | 18 | 22 | 4 | 44 | 6 |
| May | 31 | 27 | 17 | 75 | 9 |
| Total | 106 | 130 | 53 | 289 | 33 |

TABLE 21. RATIO OF ON-PROJECT SHIPMENTS TO OFF-PROJECT SHIPMENTS

| Year | On-Project | Off-Project | Total | Ratio | Cumulative Ratio |
|---|---|---|---|---|---|
| 1946 | 92 | | 92 | | |
| 1947 | 101 | | 101 | | |
| 1948 | 187 | 96 | 283 | 1.95 | 3.95 |
| 1949 | 154 | 157 | 311 | 0.98 | 2.11 |
| 1950 | 202 | 135 | 337 | 1.50 | 1.90 |
| 1951 | 285 | 137 | 422 | 2.08 | 1.95 |
| 1952 | 176 | 111 | 287 | 1.59 | 1.88 |
| 1953 | 135 | 162 | 297 | 0.84 | 1.67 |
| 1954 | 173 | 192 | 365 | 0.90 | 1.52 |
| 1955 | 214 | 292 | 506 | 0.73 | 1.34 |
| 1956* | 93 | 8 | 31 | 0.67 | 1.28 |

*Through May 20.

TABLE 22. SUMMARY OF ISOTOPE SHIPMENTS, NOVEMBER 20, 1955, THROUGH MAY 20, 1956

| Request No. | Requester | Isotope | Lot | Quantity (mg) |
|---|---|---|---|---|
| B-6151-S* | Los Alamos Scientific Laboratory | $Cr^{53}$ | GZ 870(a) | 1,000 |
| B-6152-S | Los Alamos Scientific Laboratory | $Cr^{54}$ | GZ 871(a) | 900 |
| B-5520 | Michigan State College | $Li^{7}$ | FI 668(j) | 1,000 |
| B-5966 | Massachusetts Institute of Technology | $Ge^{70}$ | BD-BE 205-210(a) | 200 |
| B-6128 | Columbia University | $Ca^{44}$ | FF 651(a) | 20 |
| B-5888* | Harvard University | $Ni^{60}$ | GT 845(a) | 1,744 |
| B-6169 | Watertown Arsenal | $Fe^{57}$ | FC 636(a) | 178 |
| B-6170 | Watertown Arsenal | $Fe^{54}$ | FC 634(a) | 177 |
| F-2780-T | Atomic Energy of Canada, Ltd. | $Ba^{138}$ | DG 435(a) | 5 |
| B-5605 | University of Arkansas | $Sr^{86}$ | HG 899(a) | 50 |
| B-5606 | University of Arkansas | $Sr^{88}$ | FL 682(a) | 50 |
| B-5607 | University of Arkansas | $Sr^{84}$ | HG 898(a) | 50 |
| B-6062 | University of Chicago | $Mo^{95}$ | AW 165(a) | 20 |
| B-6163-S | Brookhaven National Laboratory | $Sm^{147}$ | GM 814(a) | 1,000 |
| B-6164-S | Brookhaven National Laboratory | $Sm^{152}$ | GM 818(a) | 8,000 |
| B-6147 | Columbia University | $K^{41}$ | EY 626(e) | 20 |
| B-6148 | Columbia University | $Sr^{88}$ | FL 682(a) | 50 |
| B-6149 | Columbia University | $Sr^{86}$ | FL 680(a) | 10 |
| B-6190-S* | Oak Ridge National Laboratory | $Li^{6}$ | FY 745(bc) | 1,000 |
| F-2733-T | Norwegian Embassy | $Te^{128}$ | FE 646(a) | 50 |
| F-2734-T | Norwegian Embassy | $Te^{130}$ | CA 299(a) | 50 |
| F-2735-T | Norwegian Embassy | $Mo^{98}$ | R-T 58(b) | 50 |
| B-5598-S | Phillips Petroleum Co. | $Mo^{95}$ | R-T 55(a) | 382 |
| B-5602-S | Phillips Petroleum Co. | $W^{183}$ | CL 342(a) | 248 |
| B-5603-S | Phillips Petroleum Co. | $Hg^{199}$ | DR 487(ds) | 240 |
| B-6132 | U.S. Naval Radiological Defense Laboratory | $Sm^{144}$ | GM 813(b) | 50 |
| B-6187 | Massachusetts Institute of Technology | $Fe^{58}$ | GU 854(a) | 100 |
| B-6188 | Massachusetts Institute of Technology | $Fe^{57}$ | GU 853(a) | 100 |
| B-6189 | Massachusetts Institute of Technology | $Fe^{54}$ | FN 685(b) | 100 |
| F-2751-T | McMaster University | $Ba^{134}$ | FU 729(a) | 1 |
| F-2755-T | Centre National de la Recherche Scientifique (CNRS) (France) | $Hg^{202}$ | DR 490(b) | 10 |
| B-4929 | Ohio State University | $Ru^{100}$ | GJ 793(a) | 100 |
| B-5797 | Ohio State University | $Ru^{104}$ | GJ 796(a) | 15 |
| B-6089 | University of Illinois | $Pb^{204}$ | FQ 702(a) | 425 |

*Special services (see P. S. Baker and F. R. Duncan, *Stable Isotopes Research and Production Semiann. Prog. Rep. Nov. 20, 1955*, ORNL-2028, p 43).

TABLE 22 (continued)

| Request No. | Requester | Isotope | Lot | Quantity (mg) |
|---|---|---|---|---|
| B-6091 | University of Illinois | $Pb^{206}$ | BO 258(ar) | 500 |
| B-6092 | University of Illinois | $Pb^{204}$ | EO 581(a) | 470 |
| B-6141 | University of Washington | $Si^{28}$ | AI-AJ 103-106(a) | 100 |
| B-6142 | University of Washington | $Mg^{26}$ | P 49(a) | 100 |
| B-6195-S | Brookhaven National Laboratory | $Nd^{150}$ | EQ 593(ar) | 75 |
| B-6211* | University of Chicago | $Li^{6}$ | FY 745(bd) | 1,000 |
| B-6215-S | Oak Ridge National Laboratory | $K^{41}$ | DB 412(a) R | 700 |
| B-6217-S | Oak Ridge National Laboratory | $Fe^{57}$ | GU 853(a) | 200 |
| B-6220 | Massachusetts Institute of Technology | $Pd^{108}$ | GG 774(a) | 30 |
| B-6621 | Massachusetts Institute of Technology | $Cd^{116}$ | CE 321(a) | 10 |
| B-6222 | Massachusetts Institute of Technology | $Pd^{110}$ | GG 775(a) | 30 |
| B-6223 | Massachusetts Institute of Technology | $Cd^{114}$ | CE 320(ar) | 30 |
| B-6140* | University of Washington | $Si^{29}$ | GC 762(a) | 40 |
| B-6203 | University of Washington | $Mg^{24}$ | DZ 519(a) | 100 |
| B-6204 | University of Washington | $Mg^{25}$ | BZ 289(a) | 60 |
| B-6227-S | Argonne National Laboratory | $B^{10}$ | SS 1(ac) | 50 |
| B-6236-S | Brookhaven National Laboratory | $Li^{6}$ | FY 745(bd) | 5,000 |
| F-2763-T | Legation of Switzerland | $Li^{6}$ | FY 745(c) | 1 |
| F-2764-T | Legation of Switzerland | $K^{41}$ | EY 626(er) | 1 |
| F-2765-T | Legation of Switzerland | $Ag^{109}$ | EA 523(a) | 1 |
| F-2766-T | Legation of Switzerland | $Ce^{142}$ | FR 709(a) | 1 |
| F-2767-T | Legation of Switzerland | $Nd^{150}$ | EQ 593(ar) | 1 |
| F-2768-T | Legation of Switzerland | $Sm^{144}$ | FH 660(a) | 1 |
| F-2783-T | Embassy of Sweden | $Hf^{177}$ | GP 827(b) | 50 |
| B-6218 | California Institute of Technology | $Ir^{191}$ | GL 810(a) | 10 |
| B-6219 | California Institute of Technology | $Ir^{193}$ | GL 811(a) | 10 |
| B-5147* | Westinghouse Research Laboratory | $W^{182}$ | EL 562(a) | 8,000 |
| B-5148* | Westinghouse Research Laboratory | $W^{183}$ | EL 563(a) | 3,043 |
| B-5149* | Westinghouse Research Laboratory | $W^{184}$ | EL 564(a) | 8,379 |
| B-5150* | Westinghouse Research Laboratory | $W^{186}$ | EL 565(a) | 6,425 |
| B-5910* | University of Minnesota | $Fe^{54}$ | FN 685(a) | 153 |
| B-5911* | University of Minnesota | $Fe^{56}$ | DO 478(a) | 159 |
| B-6166-S | University of California Radiation Laboratory | $In^{113}$ | CQ 358(a) | 10 |
| B-6213-S | Brookhaven National Laboratory | $Ca^{48}$ | FO 694(a) | 190 |
| B-6244-S | Brookhaven National Laboratory | $Sm^{152}$ | FH 665(ar) | 300 |

*Special services.

TABLE 22 (continued)

| Request No. | Requester | Isotope | Lot | Quantity (mg) |
|---|---|---|---|---|
| B-6245-S | Brookhaven National Laboratory | $Sm^{154}$ | GM 819(a) | 200 |
| B-6246-S | Brookhaven National Laboratory | $Nd^{144}$ | EQ 589(ar) | 100 |
| B-6247-S | Brookhaven National Laboratory | $Nd^{148}$ | EQ 592(a) | 25 |
| B-6248-S | Brookhaven National Laboratory | $Nd^{146}$ | EQ 591(ar) | 200 |
| B-5802 | University of Kentucky | $Ca^{48}$ | GV 860(a) | 20 |
| B-6232* | Louisiana State University | $Ba^{130}$ | FT 720(a) | 10 |
| B-6264 | Bartol Research Foundation | $Sm^{150}$ | GM 817(a) | 100 |
| B-6266 | Bartol Research Foundation | $Sm^{144}$ | GM 813(b) | 150 |
| B-5626-S | Brookhaven National Laboratory | $Eu^{151}$ | HL 926(a) | 20 |
| B-5627-S | Brookhaven National Laboratory | $Eu^{153}$ | HL 927(a) | 20 |
| B-6241 | University of Washington | $Li^{6}$ | FY 745(bd) | 92 |
| B-6252 | University of Kentucky | $Cr^{54}$ | GZ 871(a) | 25 |
| B-6265 | Bartol Research Foundation | $Nd^{144}$ | EQ 589(ar) | 100 |
| B-6280 | Bartol Research Foundation | $Nd^{143}$ | GI 784(a) | 3,000 |
| B-6288-S | Argonne National Laboratory | $Mg^{25}$ | DZ 520(a) | 1 |
| B-6289-S | Argonne National Laboratory | $Mg^{26}$ | P 49(a) | 1 |
| B-6292-S | Phillips Petroleum Co. | $Sr^{87}$ | GH 900(a) | 1,000 |
| B-6293-S | Phillips Petroleum Co. | $Cl^{35}$ | FW 736(a) | 2,500 |
| B-6283 | Ohio State University | $Ru^{102}$ | GJ 795(a) | 165 |
| B-6297-S | Brookhaven National Laboratory | $Cu^{63}$ | DF 426(a) | 1,000 |
| F-2857-T | CNRS (France) | $Hg^{202}$ | DR 490(d) | 10 |
| B-6012-S | Argonne National Laboratory | $Sr^{87}$ | HG 900(a) | 700 |
| B-5152* | Westinghouse Research Laboratory | $Zn^{66}$ | EK 557(a) | 4,893 |
| B-5153* | Westinghouse Research Laboratory | $Zn^{68}$ | EK 559(a) | 2,833 |
| B-6282 | Johns Hopkins University | $Hg^{204}$ | DR 491(b) | 2 |
| B-6305 | University of Rochester | $Li^{6}$ | SS 5(a) | 5,000 |
| B-6319 | Cornell University | $B^{10}$ | SS 1(a) | 5,000 |
| B-6336 | University of Michigan | $Gd^{158}$ | GK 803(a) | 50 |
| B-6337 | University of Michigan | $Gd^{160}$ | GK 804(a) | 50 |
| 214 | Denmark (Dr. Nathan) | $Eu^{151}$ | HL 926(a) | 25 |
| 215 | Denmark (Dr. Nathan) | $Eu^{153}$ | HL 927(a) | 25 |
| 218 | Clarendon Laboratory (England) | $Ru^{98}$ | GJ 791(a) | 3 |
| 219 | Clarendon Laboratory (England) | $Ru^{99}$ | GJ 792(a) | 9 |
| 220 | Clarendon Laboratory (England) | $Ru^{100}$ | GJ 793(a) | 9 |
| 221 | Clarendon Laboratory (England) | $Ru^{101}$ | GJ 794(a) | 9 |

*Special services.

TABLE 22 (continued)

| Request No. | Requester | Isotope | Lot | Quantity (mg) |
|---|---|---|---|---|
| 222 | Clarendon Laboratory (England) | $Ru^{104}$ | GJ 796(a) | 9 |
| 223-S | Oak Ridge National Laboratory | $B^{10}$ | SS 1(a) | 2,400 |
| 224-S | Oak Ridge National Laboratory | $Ni^{62}$ | K 36(a) | 50 |
| 225-S | Oak Ridge National Laboratory | $Ge^{73}$ | FZ 749(a) | 50 |
| 226-S | Oak Ridge National Laboratory | $Li^{7}$ | FI 668(d) | 2,000 |
| B-5310 | Columbia University | $Te^{125}$ | HC 879(b) | 3,000 |
| 232-S | Oak Ridge National Laboratory | $B^{11}$ | FV 735(a) | 10 |
| B-6313 | American Potash & Chemical Corp. | $Li^{6}$ | HE 887(a) | 1,000 |
| B-6314 | American Potash & Chemical Corp. | $B^{10}$ | SS 1(ac) | 500 |
| B-6321 | Boston University | $B^{10}$ | SS 1(a) | 300 |
| B-6329 | University of Michigan | $Ca^{46}$ | GV 859(a) | 20 |
| B-6360-S | Brookhaven National Laboratory | $Fe^{56}$ | AZ 184(a) | 1,000 |
| B-6363 | Stanolind Oil & Gas Co. | $Sr^{86}$ | FL 680(a) | 4 |
| B-6364 | Stanolind Oil & Gas Co. | $Rb^{87}$ | GX 863(a) | 4 |
| B-6366-S | Oak Ridge National Laboratory | $Fe^{58}$ | DO 480(b) | 187 |
| 238-S | Oak Ridge National Laboratory | $Sn^{120}$ | ES 717(a) | 100 |
| B-6345-S | Brookhaven National Laboratory | $Bo^{132}$ | FU 728(a) | 10 |
| 240 | CNRS (France) | $B^{10}$ | SS 1(a) | 10,000 |
| 243-S | Oak Ridge National Laboratory | $Ni^{61}$ | RS 7(a) | 50 |
| B-6085-S* | Knolls Atomic Power Laboratory | $Li^{7}$ | FI 668(j) 1 | 942 |
| B-6086-S* | Knolls Atomic Power Laboratory | $Li^{7}$ | FI 668(j) 2 | 944 |
| B-6087-S* | Knolls Atomic Power Laboratory | $Li^{7}$ | FI 668(j) 3 | 932 |
| B-6368 | State University of Iowo | $N^{15}$ | SS 6(a) | 250 |
| 247-S | Argonne National Laboratory | $Eu^{151}$ | HL 926(a) | 50 |
| 248-S | Argonne National Laboratory | $Eu^{153}$ | HL 927(a) | 50 |
| 249-S | Oak Ridge National Laboratory | $Se^{74}$ | CY 397(ar) | 100 |
| 250-S | Oak Ridge National Laboratory | $Zn^{67}$ | RS 8(a) | 50 |
| B-6231-S* | Oak Ridge National Laboratory | $K^{39}$ | EY 624(i) | 2,100 |
| 251 | Yale University | $Mg^{25}$ | DZ 520(a) | 25 |
| 252 | Yale University | $Mg^{26}$ | BZ 290(a) | 25 |
| 253-S | Brookhaven National Laboratory | $Hf^{179}$ | CP 829(a) | 30 |
| B-6318 | Michigan State University | $Li^{6}$ | SS 5(b) | 3,000 |
| 255-S | Oak Ridge National Laboratory | $W^{184}$ | EL 564(a) | 10 |
| 256 | Naval Research Laboratory | $Ga^{71}$ | EI 555(a) | 200 |
| 257 | Naval Research Laboratory | $Rb^{87}$ | EW 620(a) | 60 |

*Special services.

TABLE 22 (continued)

| Request No. | Requester | Isotope | Lot | Quantity (mg) |
|---|---|---|---|---|
| 258-S | Phillips Petroleum Co. | $Cd^{111}$ | AC 89(a) | 238 |
| 259 | University of California | $K^{41}$ | EY 626(e) | 10 |
| 260 | Atomic Energy of Canada, Ltd. | $Li^{6}$ | SS 5(b) | 100 |
| 261 | Atomic Energy of Canada, Ltd. | $Li^{7}$ | FI 668(d) | 100 |
| 262 | Atomic Energy of Canada, Ltd. | $S^{32}$ | ES 596(a) | 20 |
| 263 | Atomic Energy of Canada, Ltd. | $Ca^{40}$ | X 70(a) | 100 |
| 264 | Atomic Energy of Canada, Ltd. | $Cl^{35}$ | FW 736(a) | 50 |
| 265 | Atomic Energy of Canada, Ltd. | $Cl^{37}$ | DH 438(a) | 50 |
| 266 | William M. Rice Institute | $Li^{6}$ | SS 5(a) | 3,000 |
| 267 | A. O. Hanson (Italy) | $Mo^{100}$ | R-T 59(br) | 40 |
| 268 | Convair – General Dynamics Corporation | $Mg^{24}$ | P 47(a) | 100 |
| 269 | Convair – General Dynamics Corporation | $Si^{28}$ | AI 103-106(a) | 100 |
| 270 | Convair – General Dynamics Corporation | $S^{32}$ | ES 596(a) | 100 |
| 271 | Convair – General Dynamics Corporation | $V^{51}$ | FM 684(a) | 100 |
| 272 | Convair – General Dynamics Corporation | $Cu^{63}$ | GT 849(a) | 100 |
| 273 | Convair – General Dynamics Corporation | $Zn^{64}$ | RS 13(a) | 100 |
| 274 | Convair – General Dynamics Corporation | $Cr^{50}$ | GR 836(a) | 100 |
| 275 | Convair – General Dynamics Corporation | $W^{186}$ | CL 344(a) | 100 |
| 276 | Massachusetts Institute of Technology | $Cd^{114}$ | CE 320(a) | 10 |
| 277 | Atomic Energy of Canada, Ltd. | $Nd^{146}$ | EQ 591(a) | 50 |
| 278-S | Westinghouse Atomic Power Laboratory | $Sm^{150}$ | EM 572(a) | 65 |
| 279-S | Westinghouse Atomic Power Laboratory | $Sm^{150}$ | FH 664(a) | 35 |
| 280-S | Westinghouse Atomic Power Laboratory | $Eu^{151}$ | HL 926(a) | 100 |
| 281-S | Westinghouse Atomic Power Laboratory | $Gd^{160}$ | GK 804(a) | 100 |
| 282-S | University of California Radiation Laboratory | $Sm^{147}$ | GM 814(a) | 50 |
| 283 | Commissariat à l'Energie Atomique (CEA) | $B^{10}$ | SS 1(a)-46 | 200,000 |
| 284 | CEA (France) | $B^{10}$ | SS 1(b) | 500,000 |
| B-5142 (285)* | Westinghouse Electric Corp. | $Ge^{70}$ | FZ 747(a) | 10,092 |
| B-5143 (286)* | Westinghouse Electric Corp. | $Ge^{72}$ | FZ 748(a) | 15,437 |
| B-5144 (287)* | Westinghouse Electric Corp. | $Ge^{73}$ | FZ 749(a) | 3,104 |
| B-5145 (288)* | Westinghouse Electric Corp. | $Ge^{74}$ | FZ 750(a) | 17,471 |
| B-5146 (289)* | Westinghouse Electric Corp. | $Ge^{76}$ | FZ 751(a) | 4,976 |
| 290 | CEA (France) | $B^{10}$ | SS 1(b) | 37,100 |
| 291 | Columbia Mineral Beneficiation Laboratory | $Ca^{44}$ | FF 651(a) | 50 |
| 292 | Anton Electronic Laboratories, Inc. | $B^{11}$ | SS 2(b) | 1,484 |

*Special services.

TABLE 22 (continued)

| Request No. | Requester | Isotope | Lot | Quantity (mg) |
|---|---|---|---|---|
| 293-S | Oak Ridge National Laboratory | $Li^6$ | SS 5(b) | 15,000 |
| 294-S | Argonne National Laboratory | $B^{10}$ | SS 1(a)-46 | 1,115,000 |
| 295 | Naval Research Laboratory | $B^{10}$ | DAG-1 | 6,000 |
| 296 | Anton Electronic Laboratories, Inc. | $B^{10}$ | DAG-1 | 5,000 |
| 297 | William M. Rice Institute | $Ni^{58}$ | GT 844(a) | 100 |
| 298 | William M. Rice Institute | $Ni^{62}$ | GT 847(a) | 10 |
| 299 | Massachusetts Institute of Technology | $Ga^{69}$ | EI 554(a) | 50 |
| 300 | CNRS (France) | $Hf^{174}$ | GP 825(a) | 2 |
| 301-S | Oak Ridge National Laboratory | $Cl^{35}$ | FW 736(a) | 50 |
| 302-S | Oak Ridge National Laboratory | $Sr^{84}$ | HG 898(a) | 10 |
| 303 | General Electric Co. | $B^{10}$ | SS 1(a)-46 | 500,000 |
| 304 | Redstone Arsenal | $B^{10}$ | SS-1(a)-46 | 10,000 |
| 305 | Redstone Arsenal | $B^{11}$ | SS 2(b) | 100,000 |
| 306 | Radiation Counter Laboratory, Inc. | $B^{10}$ | DAG-1 | 10,000 |
| 307 | Denmark (Dr. Nothan) | $Ir^{191}$ | GL 810(a) | 2 |
| 308-S | Oak Ridge National Laboratory | $B^{10}$ | SS 1(b) | 1,000,000 |
| 309 | John Hopkins University | $B^{10}$ | SS 1(b) | 10,000 |
| 310 | Iowa State College | $B^{10}$ | SS 1(b) | 10,000 |
| 311 | Naval Research Laboratory | $Ge^{73}$ | RS 29(a) | 50 |
| 312-S | Argonne National Laboratory | $S^{34}$ | GB 759(a) | 6 |
| 313 | Carnegie Institution of Washington | $Gd^{158}$ | GK 803(a) | 15 |
| 314 | Carnegie Institution of Washington | $Gd^{160}$ | GK 804(a) | 15 |
| 315-S | Argonne National Laboratory | $Hg^{204}$ | DR 491(cs) | 10 |
| 316-S | Argonne National Laboratory | $Nd^{150}$ | ER 593(ar) | 20 |
| 317-S | Argonne National Laboratory | $Zr^{96}$ | GQ 835(a) | 20 |
| 318-S | Argonne National Laboratory | $Fe^{58}$ | DL 454(f) | 100 |
| 319-S | Argonne National Laboratory | $Ca^{48}$ | GV 860(a) | 10 |
| 320-S | Argonne National Laboratory | $Pb^{208}$ | CN 352(ar) | 70 |
| 321-S | Argonne National Laboratory | $Pb^{208}$ | O 46(a) (ar) | 150 |
| 322-S | Argonne National Laboratory | $Pb^{206}$ | O 44(a) | 280 |
| 323-S | Argonne National Laboratory | $Pb^{207}$ | BO 259(a) | 200 |
| 324-S | Argonne National Laboratory | $Cr^{52}$ | HA 872(b) | 750 |
| 325-S | Brookhaven National Laboratory | $Li^6$ | SS 5(a) | 10,000 |
| 326 | National Research Council (Canada) | $Hg^{202}$ | DR 490(c) | 10 |
| 327 | Princeton University | $Mg^{25}$ | DZ 520(a) | 25 |

*Special services.

TABLE 22 (continued)

| Request No. | Requester | Isotope | Lot | Quantity (mg) |
|---|---|---|---|---|
| 328-S | Argonne National Laboratory | $Hf^{180}$ | GF 830(a) | 100 |
| 329 | University of Minnesota | $B^{10}$ | SS 1(a) | 1,000 |
| 330 | Vanderbilt University | $Sn^{112}$ | EC 527(a) | 3 |
| 331 | Vanderbilt University | $Sm^{150}$ | Em 572(a) | 10 |
| 332-S | University of California (LASL) | $Ce^{138}$ | FE 676(a) | 5 |
| 333-S | University of California (LASL) | $Ce^{140}$ | FR 708(a) | 25 |
| 334-S | Oak Ridge National Laboratory | $Cr^{54}$ | CD 312(a) | 22 |
| 335-S | Oak Ridge National Laboratory | $Fe^{57}$ | DL 453(a) | 23 |
| 337-S | Oak Ridge National Laboratory | $Zn^{67}$ | RS 15(a) | 25 |
| 338-S | Oak Ridge National Laboratory | $Zn^{70}$ | BK 344(a) (ar) | 23 |
| 339-S | Oak Ridge National Laboratory | $Ge^{73}$ | RS 29(a) | 20 |
| 340-S | Oak Ridge National Laboratory | $Ge^{76}$ | FZ 751(a) | 18 |
| 341-S | Oak Ridge National Laboratory | $Se^{77}$ | CY 399(a) | 50 |
| 342-S | Oak Ridge National Laboratory | $Se^{80}$ | FG 658(b) | 55 |
| 343-S | Oak Ridge National Laboratory | $Se^{82}$ | FG 659(a) | 30 |
| 344-S | Oak Ridge National Laboratory | $Sr^{87}$ | HG 900(a) | 50 |
| 345-S | Oak Ridge National Laboratory | $Zr^{92}$ | EE 541(a) | 50 |
| 346-S | Oak Ridge National Laboratory | $Zr^{94}$ | CV 384(a) | 40 |
| 347-S | Oak Ridge National Laboratory | $Mo^{95}$ | AW 165(a) | 30 |
| 348-S | Oak Ridge National Laboratory | $Br^{79}$ | AO 129(a) | 1 |
| 349-S | Oak Ridge National Laboratory | $Rb^{85}$ | RS 49(a) | 40 |
| 350 | Convair – General Dynamics Corporation | $B^{10}$ | SS 1(a)-46 | 20,000 |
| 351 | Rohn & Haas Co. | $B^{10}$ | SS 1(b) | 25,000 |
| 352-S | Phillips Petroleum Co. | $B^{10}$ | SS 1(b) | 5,000 |
| 353-S | Oak Ridge National Laboratory | $B^{10}$ | SS 1(a)-46 | 100,000 |
| 354-S | University of California Radiation Laboratory | $W^{184}$ | RS 56(a) | 125 |
| 355-S | University of California Radiation Laboratory | $Gd^{152}$ | GK 798(a) | 10 |
| 356-S | University of California Radiation Laboratory | $Gd^{154}$ | GK 799(a) | 30 |
| 357-S | University of California Radiation Laboratory | $Gd^{160}$ | GK 804(a) | 4.5 |
| 358-S | University of California Radiation Laboratory | $Sm^{154}$ | FH 666(ar) | 9 |
| 359 | William M. Rice Institute | $Cr^{52}$ | CD 310(a) | 100 |
| 360 | William M. Rice Institute | $Ti^{46}$ | GA 752(o) | 20 |
| 361 | Massachusetts Institute of Technology | $Hg^{199}$ | DR 487(a) | 2 |
| 362 | Massachusetts Institute of Technology | $Hg^{201}$ | DR 489(a) | 2 |
| 363 | California Institute of Technology | $Eu^{153}$ | HL 927(a) | 30 |

*Special services.

TABLE 22 (continued)

| Request No. | Requester | Isotope | Lot | Quantity (mg) |
|---|---|---|---|---|
| 364 | Neutronics Laboratory (KAPL) | $B^{10}$ | SS 1(b) | 75,000 |
| 365 | Neutronics Laboratory (KAPL) | $B^{10}$ | SS 1(b) | 20,000 |
| 366 | CEA (France) | $Mg^{25}$ | DZ 520(a) | 15 |
| 367 | CEA (France) | $Ca^{40}$ | K 70(a) | 50 |
| 368 | CEA (France) | $Cr^{52}$ | CD 310(a) | 50 |
| 369 | CEA (France) | $Sr^{88}$ | FL 682(a) | 40 |
| 370 | CEA (France) | $Cd^{112}$ | CE 318(a) | 40 |
| 371 | CEA (France) | $Cd^{113}$ | CE 319(a) | 40 |
| 372 | CEA (France) | $Cd^{114}$ | CE 320(ar) | 40 |
| 373 | CEA (France | $Sn^{116}$ | EC 530(a) | 40 |
| 374 | CEA (France) | $Sn^{118}$ | EC 532(a) | 40 |
| 375 | CEA (France) | $Sn^{120}$ | EC 534(a) | 50 |
| 376 | CEA (France) | $Ba^{138}$ | FU 733(a) | 50 |
| 377 | CEA (France) | $Pb^{208}$ | CN 352(ar) | 100 |
| 378 | CEA (France) | $Zr^{96}$ | EE 543(a) | 2 |
| 379 | CEA (France) | $Hf^{174}$ | CP 825(a) | 3 |
| 380 | CEA (France) | $Li^{6}$ | SS 5(a) | 100,000 |
| 381 | CEA (France) | $Li^{7}$ | FI 668(j) | 16,000 |
| 382 | CEA (France) | $Zn^{64}$ | RS 13(a) | 50 |
| 384 | Massachusetts Institute of Technology | $Pd^{104}$ | GG 771(a) | 30 |
| 385 | Massachusetts Institute of Technology | $Pd^{105}$ | GG 772(a) | 30 |
| 386 | Massachusetts Institute of Technology | $Pd^{106}$ | GG 773(a) | 30 |
| 387 B-5151* | Westinghouse Electric Corp. | $Cr^{52}$ | GZ 869(a) | 27,012 |
| 388 B-5154* | Westinghouse Electric Corp. | $Ti^{46}$ | GA 752(a) | 3,638 |
| 389 B-5155* | Westinghouse Electric Corp. | $Ti^{47}$ | GA 753(a) | 2,906 |
| 390 B-5155* | Westinghouse Electric Corp. | $Ti^{47}$ | GA 753(b) | 2,570 |
| 391 B-5156* | Westinghouse Electric Corp. | $Ti^{48}$ | GA 754(a) | 37,480 |
| 392 B-5157* | Westinghouse Electric Corp. | $Ti^{49}$ | GA 755(b) | 4,128 |
| 393 B-5158* | Westinghouse Electric Corp. | $Ti^{50}$ | GA 756(a) | 1,402 |
| 394 B-5158* | Westinghouse Electric Corp. | $Ti^{50}$ | GA 756(b) | 1,578 |
| 395 | Brookhaven National Laboratory | $Sm^{154}$ | GM 819(a) | 50 |
| 396 | Oak Ridge National Laboratory | $Hf^{177}$ | GP 827(a) | 10 |
| 397 | Oak Ridge National Laboratory | $Hf^{179}$ | GP 829(b) | 10 |
| 398 | Oak Ridge National Laboratory | $Hf^{180}$ | GP 830(a) | 10 |
| 399 | Massachusetts Institute of Technology | $B^{10}$ | SS 1(a) | 1,000 |

*Special services.

TABLE 22 (continued)

| Request No. | Requester | Isotope | Lot | Quantity (mg) |
|---|---|---|---|---|
| 400 | Argonne National Laboratory | $W^{180}$ | HN 931(a) | 5 |
| 401 | Washington University | $B^{10}$ | SS 1(a) | 1,000 |
| 402 | Brookhaven National Laboratory | $Hf^{180}$ | GP 830(a) | 20 |
| 403 | Bartol Research Foundation | $Se^{74}$ | BS 268(a) | 10 |
| 405 | Brookhaven National Laboratory | $Gd^{152}$ | GK 798(a) | 3 |
| 406* | University of Minnesota | $Cd^{111}$ | RS 61(a) | 89 |

*Special services.