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STABLE ISOTOPE RESEARCH AND PRODUCTION

DIVISION SEMIANNUAL PROGRESS REPORT

FOR PERIOD ENDING NOVEMBER 20, 1953

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STABLE ISOTOPE RESEARCH AND PRODUCTION DIVISION SEMIANNUAL PROGRESS REPORT

for Period Ending November 20, 1953

C. P. Keim, Director

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STABLE ISOTOPE RESEARCH AND PRODUCTION DIVISION SEMIANNUAL PROGRESS REPORT

ISOTOPE ANALYSIS METHODS LABORATORY

J. R. McNally, Jr., Department Head

INTRODUCTION

The work of the Isotope Analysis Methods Laboratory is in the field of spectroscopic investigations of elements and isotopes and their compounds. Specific fields of investigation covered are nuclear resonance, microwave and infrared spectroscopy, optical spectroscopy, spectrochemistry, and x-ray absorption and diffraction. The work is reported on a project basis to give a more complete picture of the purpose, activity, and status of each project. More detailed information on these projects may be obtained from the previous semiannual report.[1]

NUCLEAR RESONANCE

H. E. Walchli

Project NR-1 – Table of Nuclear Moment Data

The general objective of Project NR-1 is to collect all pertinent information on the nuclear magnetic moments of nuclides. A compilation[2] of such data and a supplement[3] have been issued in tabular form. These tables contain data on nuclear magnetic resonance frequencies, nuclear spins, measurement methods, frequency ratios determined, chemical compounds used, and precision of measurement. In addition, working tables of diamagnetically corrected nuclear magnetic moments and standardized frequency ratios have been included, as well as a discussion of chemical shielding effects. Supplement No. 2 is in preparation and will be issued early in 1954.

Project NR-2 – Nuclear-Resonance Analytical Applications

Project NR-2 was temporarily inactive so as to permit equipment changes (cf. "Project NR-3," this report). A new head unit for HF studies has been constructed by using silver-plated components and Teflon coil supports.

Project NR-3 – Nuclear Magnetic Resonance Equipment Development

A precision resistor for controlling the magnetic field has been installed, and the regulator amplifier has been rebuilt and will give a system transconductance, $\Delta I/\Delta V$, of 65 mhos. Magnetic-field control is now such that a variation of 50 μv in the input produces a change of 10^{-2} gauss in field at field strengths ranging up to 10^4 gauss. A short-time regulation of 1 part in 10^5 is obtained, but a long-time drift of 1 part in 10^4 is expected because of the use of a direct-coupled amplifier. This amplifier is to be replaced shortly with a high-gain system that uses a chopper and an a-c amplifier.

A shielded room giving an attenuation of 100 db from 100 kc to above 1000 megacycles has been installed around the nuclear-resonance equipment. Tests to date indicate complete elimination of interference from short-wave broadcast stations which, in the past, have given considerable measurement difficulties. In addition, all noise from other laboratory equipment has been substantially eliminated.

Project NR-4 – Measurement of Nuclear Magnetic Moments

Equipment modification and shielding installation have prevented measurements during the first five months of this report period. Following completion of this work, remeasurements of nuclear-resonance frequency ratios were made on nine nuclides. Detection, recognition, and measurement of these frequencies have been markedly improved as a direct result of this equipment development.

---

[1] *Stable Isotope Research and Production Division Semiann. Prog. Rep. May 20, 1953*, ORNL-1563, p. 1.

[2] H. E. Walchli, *A Table of Nuclear Moment Data*, ORNL-1469 (April 1, 1953).

[3] H. E. Walchli, *A Table of Nuclear Moment Data, Supplement No. 1*, ORNL-1469 (June 1, 1953).

MICROWAVE SPECTROSCOPY

Project M-1 – Microwave Spectrum of Vinyl Fluoride

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

Measurements of the microwave spectrum of vinyl fluoride, CH_2:CHF, are completed for the $J = 0$ to 1 and $J = 1$ to 2 transitions. Final values of the rotational constants and of the molecular asymmetry parameter are:

a = 37,515.55 ± 1.00 megacycles,
b = 10,621.95 ± 0.04 megacycles,
c = 9,103.07 ± 0.04 megacycles,
K = –0.89304 ± 0.00001,

where

$$a = \frac{\hbar^2}{2I_a}, \quad b = \frac{\hbar^2}{2I_b}, \quad \text{and} \quad c = \frac{\hbar^2}{2I_c}.$$

Previously, K had been estimated at –0.95. Computations of the electric dipole moment components are being completed.

Project M-2 – Microwave Spectrum of Vinyl Acetylene

H. W. Morgan J. H. Goldstein

Measurements on 15 microwave transitions arising from the $J = 3$ to 4 and $J = 4$ to 5 rotational levels have been completed. The 15 lines have been expressed as the sum of seven terms (eight unknowns including the asymmetry parameter δ) as follows:

$$\nu = (a + c)(J + 1) + \left(\frac{a - c}{2}\right)E_{J\tau}(\delta) + A_1(W_0'^2 - W_0^2)$$
$$+ A_2\left[W_0'J'(J' + 1) - W_0J(J + 1)\right] + A_3\left[J'^2(J' + 1)^2 - J^2(J + 1)^2\right]$$
$$+ A_4\left[K^2J'(J' + 1) - J(J + 1)\right] + A_5K^2(W_0' - W_0),$$

where ν is the radiated frequency, the A's are centrifugal distortion constants, $E_{J\tau}(\delta)$ is a function of the asymmetry, and the W's, J's, and K's are the energies and quantum numbers for the upper (′) and lower states. In this relation, the first two terms involve the moments of inertia, and the last five terms involve centrifugal distortion of the molecule. Least-squares solution of the data on vinyl acetylene is being carried out by the Mathematics Panel.

Project M-3 – Microwave Spectrometer Equipment Development

H. W. Morgan

The acquisition of several extremely sensitive 1N53 crystals for microwave radiation detectors has improved the sensitivity to such a degree that a 100-kc preamplifier is no longer required. Sensitivity is better by as much as a factor of 10 over previously used 1N26 crystals and, in addition, noise appears to be lower by at least a factor of 2.

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

INFRARED SPECTROSCOPY

Project IR-1 – Infrared Spectra of Lithium Alkyls

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

Preliminary infrared studies in which petroleum ether was used as the solvent have been made of the lithium alkyls. A new technique involving the use of infrared, transparent, "vacuum-pressed" KBr windows as a matrix for solid samples permits the infrared spectra of the samples to be obtained free from any background spectra. Solid alkyls are to be examined by using this technique. No significant data have yet been obtained.

Project IR-2 – Infrared and Raman Spectra of $LiAlH_4$ and $LiAlD_4$

P. A. Staats J. H. Goldstein

The primary objectives of Project IR-2 are to obtain infrared and Raman spectra of $LiAlD_4$ and to study isotope shifts in the fundamental vibration frequencies of the AlH_4^- and AlD_4^- ions. Also, it is hoped to ascertain whether the structure is or is not tetrahedral. The four fundamental

vibrations for $LiAlH_4$ have been reported by Lippincott.[5] In the course of synthesizing deuterated lithium isopropyl, $(CH_3)_2CDLi$, for Project IR-1, it was necessary to prepare $LiAlD_4$; consequently, infrared studies of this molecule were considered to be in order.

Two $LiAlD_4$ infrared active fundamentals (ν_3 = 1284 cm^{-1} and ν_4 = 563 cm^{-1}) have been assigned. Raman spectrum studies, in which equipment at the University of Tennessee[6] was used, have been unsuccessful to date. Attempts to synthesize $LiAlH_3D$ and $LiAlHD_3$ have been unsuccessful. Subsequent work will involve the determination of the ν_1 and ν_2 fundamentals in the Raman spectrum and the computation of the AlH_4^- and AlD_4^- structure.

Project IR-3 – Infrared and Microwave Spectra of Formyl Fluoride

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

A study of the molecules HCOF and DCOF is directed toward the evaluation of the molecular parameters and the computation of thermodynamic properties. The formyl fluoride molecule is a planar, near-symmetric-top molecule with six fundamental vibrations expected. This compound is of interest to the ORNL organic chemists who have furnished two samples of HCOF, one from a C^{14} radioactive tracer experiment.

The six fundamental vibrations of HCOF in the infrared region have been observed, and assignments have been made (see Table 1). The two C-H bending vibrations are extremely weak. The resolved infrared fine structure indicates good agreement with the tentative molecular rotation parameters based on the observed microwave spectra. The electric dipole moment of the molecule appears to be closely parallel to the *b* axis; microwave assignments are yet to be made. Work is progressing on the preparation of DCOF, and the isotope shifts in both the infrared and microwave regions will be measured, and assignments will be confirmed when the preparation is completed.

[5]E. R. Lippincott, *J. Chem. Phys.* 17, 1351 (1949).

[6]The authors are indebted to Prof. A. H. Nielsen for the use of the Raman spectrograph.

TABLE 1. FUNDAMENTAL VIBRATIONS OF THE FORMYL FLUORIDE MOLECULE

VIBRATION	TYPE OF VIBRATION	FREQUENCY (cm^{-1})
C-H	Stretching	2954
C-O	Stretching	1838
C-F	Stretching	1062
C-H	Bending-parallel	1021
C-H	Bending-perpendicular	822
FCO	Bending	659

OPTICAL SPECTROSCOPY

Project OS-1 – Plane-Grating Monochromator

G. K. Werner

The grating mounting of the monochromator has been designed and is now being fabricated. Mirror mountings have been installed on the 16-ft lathe bed, and readjustments of the slit assembly have been made. Various spectrum receivers to be introduced in the exit plane are in the design stage.

Project OS-2 – Spectroisotopic Assay of Lithium

D. D. Smith Z. Combs

Continued developmental work on the lithium assay technique by optical spectroscopy has resulted in several significant improvements. In October, better optical filtering of spurious lines and of background light resulted in considerably higher reproducibility. The relative error introduced by spectrum drift in the sampling of various standards is indicated in Table 2 (in general, the peak is set to within ±4 mA but may drift as much as 20 mA during a protracted measurement period). Table 3 indicates the precision obtained at various abundance levels in individual 20-sec measurements, and Table 4 illustrates the reproducibility of sampling. Sampling rates as high as five assays per hour have been attained which indicate the high speed of spectroscopic measurements. Samples as small as 100 μg of lithium have been successfully analyzed. A projected application of this lithium assay technique is to a more simplified, routine assay method for uranium. Preliminary uranium studies are under way.

TABLE 2. BIAS DUE TO LINE-PEAKING ERRORS IN ASSAY OF LITHIUM

ERROR IN LINE-PEAKING (mA)	PERCENTAGE OF BIAS AT VARIOUS ABUNDANCE LEVELS			
	At 13%	At 30%	At 59%	At 76%
+20	+0.8	+0.9	+0.4	−0.2
+10	+0.3	+0.3	+0.2	+0.2
0	0.0	0.0	0.0	0.0
−10	−0.3	0.0	+0.2	−0.2
−20	+0.8	+0.3	−0.2	0.0

TABLE 3. WITHIN-SAMPLE PRECISION AT VARIOUS Li^6 ABUNDANCE LEVELS

MEASUREMENT NUMBER	PERCENTAGE OF Li^6 AT VARIOUS ABUNDANCE LEVELS				
	Normal	At 10%	At 20%	At 30%	At 80%
1	7.30	9.66	20.41	30.28	77.47
2	7.21	9.80	20.41	30.44	77.43
3	7.30	9.90	20.26	30.44	78.41
4	7.12	9.83	20.50	30.59	77.84
5	7.30	9.90	20.54	30.12	78.30
6	7.36	9.70	20.41	30.28	78.04
7	7.12	9.80	20.11	30.28	77.95
8	7.15	9.80	20.26	30.28	77.62
9	7.39	9.90	20.41	30.22	78.04
10	7.39	9.70	20.21	30.12	77.58
Average	7.26	9.80	20.35	30.31	77.87
Maximum limit of error	0.24	0.20	0.31	0.33	0.76
Average limit of error	0.077	0.064	0.099	0.109	0.24
Percentage limit of error of Li^6	1.06	0.65	0.49	0.35	0.31

Project OS-3 – Photodetector Investigations

O. B. Rudolph G. K. Werner

Facilities for testing multiplier phototubes have been completed. A spacious light-proof room with provisions to introduce light of variable wavelength, intensities, and apertures is being used in studies of phototubes at near-noise-level conditions and close to wavelength-sensitivity "cut-off." A figure of merit being used at present is the luminous equivalent of the background noise. These preliminary investigations of tubes have indicated small errors in the measure and in the record cycles of a Baird Associates Direct Reader and should lead to improved light-measurement techniques.

Project OS-4 – Helium Spectra

O. C. Yonts K. L. Vander Sluis

An atomic beam source for measuring helium spectra has been constructed and will be tested shortly.

TABLE 4. REPLICATE SAMPLING OF LITHIUM STANDARDS

25.25% STANDARD			30.15% STANDARD		
Sample	Abundance (%)	Error (%)	Sample	Abundance (%)	Error (%)
Control	(25.25)	(0.00)	Control	(30.15)	(0.00)
1	25.20	−0.05	1	30.36	+0.21
2	25.40	+0.15	2	30.03	−0.12
3	25.35	+0.10	3	30.58	+0.43
Control	(25.25)	(0.00)	Control	(30.15)	(0.00)
4	25.36	+0.11	4	29.94	−0.21
5	25.22	−0.03	5	30.07	−0.08
6	25.39	+0.14	6	29.92	−0.23
Control	(25.25)	(0.00)	Control	(30.15)	(0.00)
7	25.23	−0.02	7	30.17	+0.02
8	25.22	−0.03	8	30.15	0.00
9	25.15	−0.10	9	30.50	+0.35
Control	(25.25)	(0.00)	Control	(30.15)	(0.00)
10	25.30	+0.05	10	30.38	+0.23
11	25.50	+0.25	11	29.86	−0.29
12	25.45	+0.20	12	30.04	−0.11
Control	(25.25)	(0.00)	Control	(30.15)	(0.00)
13	25.10	−0.15	13	30.11	−0.04
14	25.20	−0.05	14	30.07	−0.08
15	25.05	−0.20	15	29.93	−0.22
Control	(25.25)	(0.00)	Control	(30.15)	(0.00)
Limit of error = 0.285%			Limit of error = 0.47%		
Percentage limit of error of Li^6 = 1.13%			Percentage limit of error of Li^6 = 1.56%		

Project OS-6 – Hyperfine Structure of U^{233} and U^{235}

K. L. Vander Sluis

The nuclear spin of U^{233} has been determined to be $I = \frac{5}{2}$ (see Fig. 1). The magnetic moment is opposite that of U^{235}, while the U^{233} nuclide has a quadrupole moment comparable to that of U^{235}. Although there are numerous resolvable hyperfine patterns in U^{233}, none have yet been found in U^{235} because of the quadrupolar compression of levels.

Excitation of uranium spectra by use of UCl_4 in a discharge tube and a Raytheon 2450-megacycle diatherm unit has been successfully produced; the excitation is predominantly atomic. To enhance the efficiency of the Fabry-Perot interferometer, monochromatic light (e.g., λ 5915 A of uranium) is obtained by using a high-dispersion spectrograph for premonochromatization; thus, entrance-aperture limitation rather than exit-aperture limitation is obtained at the interferometer.

Project OS-7 – Zeeman Spectroscopy

P. M. Griffin

Satisfactory sources for d-c arc and for low-power spark excitation of uranium in the magnetic

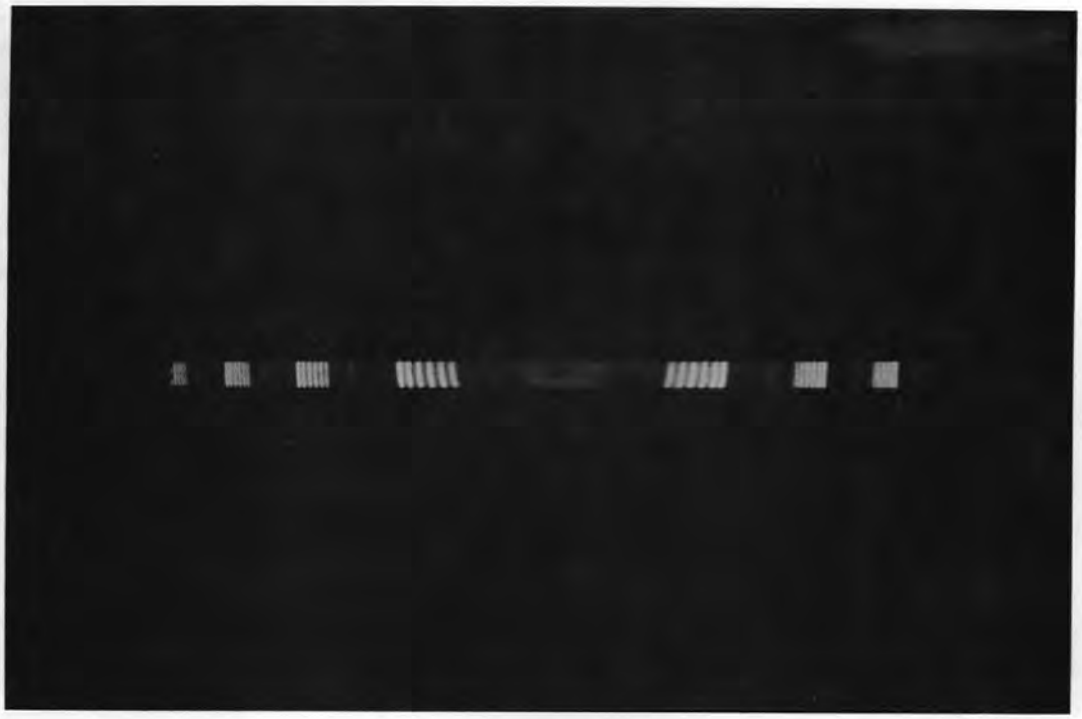

Fig. 1. Fabry-Perot Hyperfine Pattern of U^{233} Showing Six-Component Pattern ($I = \frac{5}{2}$).

field have been developed. Zeeman spectra of uranium up to 8900 A have been photographed at a field strength of 25,000 gauss. A low-field (10,000 gauss) magnet used for nuclear resonance work has been tested with a uranium electrodeless discharge tube, and results indicate the feasibility of inserting pole pieces to boost the field to above 30,000 gauss.

Project OS-8 – Theory of the Aperture-Limited Fabry-Perot Interferometer

K. L. Vander Sluis

The problem associated with the use of the aperture-limited Fabry-Perot interferometer has been considered by Tolansky[7] and has been treated in more detail by Geiger.[8] A review of this work indicates an erroneous theoretical approach which affects the final expressions. In particular, the interferometer must be considered as an instrument which produces interference by division of amplitude, not by division of wave front, although the latter may occur at certain larger angles, depending on the degree of coherence in the wave front.

Expressions have been developed for three cases of importance, namely, exit aperture equal to, larger than, or less than the entrance aperture. It is concluded that the optimum situation occurs for exit aperture larger than the entrance aperture, subject, of course, to the quality and flatness of the interferometer plates. The condition for using an interferometer at 99% efficiency is

$$n = \frac{1}{\log R} \log \left[\frac{(1 + R)(1 - R^k)}{1 - R^{2k}} - \sqrt{\frac{(1 + R)^2 (1 - R^k)^2}{(1 - R^{2k})^2} - \frac{0.01k(1 - R^2)}{1 - R^{2k}}} \right],$$

where $n \cong (A - a)/2t\theta$ is the number of reflections the ray can have beyond the entrance aperture ($2a$) before being cut off by the exit aperture ($2A$), $k \cong a/t\theta$ is the number of reflections the ray may

[7] S. Tolansky, *High Resolution Spectroscopy*, p. 161, Methuen & Co., ltd., London, 1947.

[8] F. E. Geiger, Jr., *J. Opt. Soc. Amer.* 39, 249 (1949).

have in traveling a distance across the interferometer equal to the length of the entrance aperture, and R is the reflection coefficient. As an example, with 95% reflectivity, a 20-mm spacer, and a required five rings for interpretation, it can be computed that an exit aperture of at least 10 cm is required.

The effective k beyond which no n is required (matched apertures) can be obtained when the bracketed expression is set equal to zero, and then k will be approximately 3000, or the entrance aperture ($2a$) must be 150 cm! If only an 80% efficient interferometer is required for the above case, then k must be only 150 and $2a \cong 7$ cm.

SPECTROCHEMISTRY

Project SC-1 – General Spectrochemical Analysis

M. R. Skidmore J. A. Norris
E. L. Grove[9]

New general standards for spectrochemical analysis in a graphite matrix have been prepared, with minor changes in element groupings and concentrations. A survey of alkali suppression and enhancement effects has been made.

Project SC-2 and SC-3 – Rare-Earth Stainless Steels and Other Reactor Materials

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

Minor changes in the standards for rare-earth stainless steels and other reactor materials have been introduced.

X-RAY ANALYSIS

Project X-1 – Chemical Analysis by X-Ray Absorption and Fluorescence

W. F. Peed H. W. Dunn

Activity in the field of chemical analysis by x-ray absorption and fluorescence now includes the study of rare earths and of oxide films on stainless steels. Fluorescence L spectra of relatively pure samarium, gadolinium, terbium, dysprosium, erbium, and ytterbium and the K spectra of yttrium and chromium have been recorded, with chromium to be used as a reference standard. Analysis lines as free from other rare-earth interferences as is possible have been selected. Intensity ratios between these selected lines and the line of chromium K_α are being obtained for various concentrations to establish working curves. Preliminary x-ray fluorescence studies of oxide films on stainless steels used in reactor experiments indicate that x-ray investigations may assist in this problem.

Project X-2 – X-Ray Diffraction Analysis

H. W. Dunn

Work is continuing on the tabulation of x-ray diffraction data for over 100 compounds studied in this laboratory for which no information is available in either the classified literature or in published reports. To date, calculations have been completed on 27 compounds. Of particular interest to x-ray laboratories holding AEC contracts are the data on lithium, sodium, and potassium fluochromates which are corrosion products of the hot, alkali fluoride heat-exchange media on stainless steels. Phase diagram studies are nearly complete.

Project X-3 – Isotope Effects in X-Ray Spectra

W. F. Peed G. L. Rogosa[10]

The objective of cooperative project X-3 is to investigate the possibility of isotope effects in the L_I level of uranium. Previous work[11] indicates no isotope effects in the L_{II} and L_{III} levels of U^{235} and U^{238}. The limit of detectability is believed to be about 0.4 volt or 3000 cm^{-1}. Samples of U^{233}, U^{235}, and U^{238} prepared as the oxide in pressed copper powder will be soldered to the x-ray target and bombarded with high-energy electrons. Direct excitation of the uranium should enhance the detectability of any structure in the L_I transitions. Rough estimates of the effect expected are close to this limit of detectability.

SPECIAL PROJECTS

Project SP-2 – Automatic Data Accumulation

W. F. Peed H. E. Walchli

Components of a prototype automatic data accumulator (ADA), designed for application to x-ray and photoelectric analysis problems, have been

[9] Summer research participant, University of Alabama, University, Alabama.

[10] Former research participant, Florida State University, Tallahassee, Florida.

[11] G. L. Rogosa and G. Schwarz, unpublished material, 1953.

tested. The present operation schedule of the ADA includes (1) pre-exposure at controlled intensity for any number of operations to a maximum of nine, (2) exposure and measure for any number of operations up to a maximum of eight, and (3) the recording of two time intervals to 0.01 sec and a current (or voltage) to two significant figures.

Project SP-3 – Arc-Discharge Problems

O. C. Yonts

Studies of arc filaments used in calutron discharges have established the relation between filament erosion and arc power[12] in the calutron to be $e = kP^{3/5}$. Unusual activation of tantalum filaments by rare-earth charge materials has been shown to be due to a coating of gadolinium metal. Above 1650°C, emission reverts to that characteristic of pure tantalum. The work function of gadolinium-activated tantalum has been evaluated as 3.6 volts.

[12] C. H. Townes, *Phys. Rev.* 65, 319 (1944); a discussion of the general problem of cathode sputtering.

TABLE 5. TANTALUM-183 GAMMA-RAY CLASSIFICATIONS

LINE	INTENSITY*	ENERGY (kev)*	TRANSITION	CALCULATED ENERGY (kev)	ERROR
A		46.481 ± 0.005	12-10	46.48	0.00
B		52.593 ± 0.005	10-8	52.59	0.00
C	Weak	83 ± 1	7-4	81.40	−2
			7-3	82.28	−1
D	1.4	99.070 ± 0.010	12-8	99.07	0.00
E	2.4	107.933 ± 0.012	8-6	107.94	+0.01
F	0.4	109.729 ± 0.013	8-5	109.74	+0.01
G	0.1	110.395 ± 0.150	9-7	110.41	0.01
H	0.4	143.773 ± 0.050	8-4	143.77	0.00
I	2.7	144.151 ± 0.050	2-1		
J	1.1	160.532 ± 0.030	10-6	160.53	0.00
K	3.3	161.458 ± 0.025	12-7	161.44	−0.02
L	1.8	162.335 ± 0.030	10-5	162.33	−0.01
M	0.6	192.692 ± 0.065	9-3	192.69	0.00
N	1.8	205.062 ± 0.050	13-8	205.09	+0.03
O	1.9	209.823 ± 0.035	3-1 ?	209.34	−0.48
			8-2	209.84	+0.02
			4-1 ?	210.22	+0.40
P	Weak	236 ± 1	11-4	236	0
			11-3	237	1
Q	3.0	244.259 ± 0.040	5-1	244.25	−0.01
R	10	246.051 ± 0.030	6-1	246.05	0.00
S	2.5	291.708 ± 0.060	7-1	291.62	−0.09
T	2.6	313.078 ± 0.065	13-6	313.03	−0.05
U	3.9	354.038 ± 0.090	8-1	353.99	−0.05

*See footnote 13, this section.

Project SP-4 – Vacuum Line for Gas Handling

K. L. Vander Sluis P. M. Griffin
H. W. Morgan

A vacuum rack for the handling and the preparation of gas samples for microwave, infrared, and optical studies of discharge tubes used in spectroscopic work has been completed.

Project SP-5 – Decay Scheme for Ta^{183}

J. R. McNally, Jr.

A decay scheme for Ta^{183} has been proposed which is based on the excellent gamma-ray energies reported by DuMond *et al.*[13] Thirteen gamma-ray levels are established, only four of which are based on less than three transitions. Agreement for the nine definitely established levels is well within the reported precision. Tables 5 and 6 summarize the data.

TABLE 6. PROPOSED GAMMA-RAY-LEVEL SCHEME FOR TANTALUM-183

LEVEL REFERENCE	ENERGY (kev)	NUMBER OF TRANSITIONS
1	0.00	5
2	144.15	2
3	209.34	3
4	210.22	3
5	244.25	3
6	246.05	4
7	291.62	4
8	353.99	8
9	402.03	2
10	406.58	4
11	446	1
12	453.06	3
13	559.08	2

PUBLICATIONS

Reports and papers in preparation or submitted for publication:

1. K. L. Vander Sluis and J. R. McNally, Jr., "Nuclear Moments of U^{233} and U^{235}," to be published in the *Journal of the Optical Society of America.*
2. K. L. Vander Sluis and J. R. McNally, Jr., "Fabry-Perot Interferometer with Finite Apertures."
3. E. L. Grove and J. A. Norris, "The Effects of Certain Alkali Compounds in the Burning of Powdered Graphite Samples."
4. E. L. Grove, "The Effect of Extraneous Elements on Spectral Line Intensities."
5. H. W. Morgan and J. H. Goldstein, "Second-Order Quadrupole Effects in Asymmetric Tops: The Microwave Spectrum of Vinyl Iodide."
6. H. W. Morgan and J. H. Goldstein, "The Microwave Spectrum and Molecular Structure of Vinyl Fluoride."
7. H. E. Walchli, "A Table of Nuclear Moment Data," Supplement No. 2.
8. O. C. Yonts, W. A. Bell, and L. O. Love, "Preliminary Report on Filament Activation with Gadolinium."
9. O. C. Yonts, "Filament Life in Low Pressure Arc Type Ion Source."

Reports published and papers presented:

1. G. K. Werner, "Semi-Automatic Recording of Wavelength Data," *J. Opt. Soc. Amer.* **43**, 620 (1953).
2. J. R. McNally, Jr., "Spectroscopic Research at Oak Ridge National Laboratory," presented at meeting of the Cleveland Society of Spectroscopy, Cleveland, Ohio, September 23, 1953.
3. J. R. McNally, Jr., "Atomic Spectroscopy of the Nucleus," presented at the ORINS-ORNL Fifth Annual Oak Ridge Summer Symposium, August 24-29, 1953.
4. H. E. Walchli, *A Table of Nuclear Moment Data, Supplement No. 1*, ORNL-1469 (June 1, 1953).

[13] J. W. M. DuMond, H. C. Hoyt, P. E. Marmier, and J. J. Murray, *Phys. Rev.* **92**, 202 (1953).

MASS SPECTROMETER LABORATORY

Russell Baldock, Department Head

INTRODUCTION

Extensive investigations have been made on the uranium content of aircraft fuels. By employing the isotope-dilution technique, quantitative measurements with a routine accuracy of ±1% have been made on the fuel burnup. Considerable effort has been expended in attempting to develop a rapid and accurate method of determining small amounts of nickel and large amounts of uranium in the presence of high fission-product activity. As was anticipated, the efficient generation of nickel ions is difficult to achieve. Preliminary results indicate that no difficulty will be experienced in obtaining high-precision analyses of the uranium content of Chattanooga shale. New determinations have been completed on the isotopic abundance of natural palladium, and theoretical investigations have been initiated on the constitution of metal halide vapors.

INVESTIGATIONS OF ARE FUELS

Russell Baldock	H. K. Gladden[2]
H. E. Carr[1]	R. E. Rummel[3]
L. O. Gilpatrick	J. R. Sites

The mass spectrometer has been successfully employed in the quantitative analysis of the uranium content of aircraft fuels. Initially, the principal effort was directed toward testing the validity of chemical analyses which were then in question. These difficulties were soon resolved, and it was recognized that the methodology could readily be extended to obtain an accurate measure of the fuel burnup. In addition, by using accepted values of cross sections, an average value of the flux in which the fuel has been irradiated can be determined. This gives an independent check with predicted values based on other types of measurements. Thus, the internal consistency of the calculations can be established.

Certain key features of the isotope-dilution method, as applied to the analysis of the aircraft fuel, are contained in the following paragraphs. The method has been used extensively and gives a routine accuracy of ±1%, but this is not the ultimate limit of accuracy. The ratio, R, as subsequently defined, is the particular quantity that is measured directly by the mass spectrometer. An outstanding feature of this method of analysis is that the chemistry of sample preparation needs to be quantitative in only the initial step, which is that of putting the fuel and the spike in solution.

Calculation of UF_4 in Unirradiated Fuels. The procedure given below is used for calculating the percentage by weight of UF_4 in a fuel sample of known isotopic composition. By use of the exact values of the isotopic masses, the atomic weight, M_f, of the uranium in the fuel is computed as follows:

$$M_f = 235.12\,A_{235} + 234.12\,A_{234} + 238.12\,A_{238}\,,$$

where A_n denotes abundance of uranium isotope of mass number n. The ratio, R, of U^{235} to U^{238} is obtained from the mass spectrometer measurement, and since the atoms of U^{235} and U^{238} are obtained from both the uranium in the fuel and the natural U_3O_8 spike material added,

$$R = \frac{(\text{atoms of } U^{235} \text{ from fuel}) + (\text{atoms of } U^{235} \text{ from spike})}{(\text{atoms of } U^{238} \text{ from fuel}) + (\text{atoms of } U^{238} \text{ from spike})}\,.$$

The atomic weight of uranium in the spike is 238.07; therefore,

$$(1) \qquad R = \frac{\dfrac{W_1 Y F_g A_{235}}{M_f} + \dfrac{W_2 F_g^* A_{235}}{238.07}}{\dfrac{W_1 Y F_g A_{238}}{M_f} + \dfrac{W_2 F_g^* A_{238}}{238.07}}\,,$$

[1]Consultant, Alabama Polytechnic Institute.

[2]Research participant, Georgia Institute of Technology.

[3]Consultant, Vanderbilt University.

where

W_1 = weight of uranium-bearing fuel,

W_2 = weight of spike (U_3O_8),

F_g = U/UF_4 = gravimetric factor for uranium in fuel,

F_g^* = $3U/U_3O_8$ = gravimetric factor for uranium in spike,

Y = weight fraction of UF_4 in fuel.

If Eq. 1 is solved for Y,

$$(2) \qquad Y = \frac{\dfrac{W_2F_g^*A_{235}}{238.07} - R\,\dfrac{W_2F_g^*A_{238}}{238.07}}{R\,\dfrac{W_1F_gA_{238}}{M_f} - \dfrac{W_1F_gA_{235}}{M_f}}$$

and $Y \times 100$ = percentage of UF_4, by weight, in the sample.

Calculation of U^{235} Lost from Irradiated Fuels. The procedure given below is used for calculating the percentage of U^{235} lost from irradiated fuel spiked with natural U_3O_8. If C is defined as the number of gram atomic weights of U^{235} in W_1 grams of fuel before irradiation, then

$$(3) \qquad C = \frac{W_1YF_gA_{235}}{M_f} ,$$

and, for the irradiated spiked-fuel sample, the following equation must be true:

$$(4) \qquad R = \frac{X + \dfrac{W_2F_g^*A_{235}}{238.07}}{\dfrac{W_1YF_gA_{238}}{M_f} + \dfrac{W_2F_g^*A_{238}}{238.07}} ,$$

where X is the number of gram atomic weights of U^{235} present in W_1 grams of fuel after irradiation. Solving Eq. 4 for X gives

$$(5) \qquad X = R\,\frac{W_1YF_gA_{238}}{M_f} + R\,\frac{W_2F_g^*A_{238}}{238.07} - \frac{W_2F_g^*A_{235}}{238.07} .$$

Now $C - X$ = the number of gram atomic weights of U^{235} lost from W_1 grams of fuel; therefore, the percentage of U^{235} lost from the fuel is

$$\left(1.00 - \frac{C - X}{C}\right) 100 .$$

Equation 5, however, gives only the total percentage of U^{235} lost from the fuel. This loss may be due to several factors, including burnup, radiation damage, and the replacement of chromium leached from the Inconel walls. A graphical method of testing has been developed which will be used to determine whether the loss can be completely ascribed to the capture of neutrons and the subsequent production of fission products and U^{236} (i.e., to burnup).

The solution of the differential equation for the first-order rate expression for burnup is

$$(6) \qquad \log \frac{U_0^{235}}{U_t^{235}} = \frac{(\alpha_1 + \alpha_2)\phi^t}{2.303} ,$$

where

α_1 = fission cross section = 580 barns,

α_2 = capture cross section = 105 barns,

ϕ = neutron flux density = 2.17×10^{14} neutrons/cm^2· sec,

t = irradiation time, sec,

and the slope is

$$(7) \qquad \frac{(\alpha_1 + \alpha_2)\phi}{2.303} = 6.4544 \times 10^{-8} ;$$

therefore,

$$(8) \qquad \log \frac{U_0^{235}}{U_t^{235}} = 6.4544 \times 10^{-8}t .$$

If a plot is made of Eq. 8, which might be called the theoretical curve, it is possible to compare the experimental results on burnup with the results predicted by theory. In addition, Eq. 8 can be used to check the validity of the products $\alpha_1\phi$ and $\alpha_2\phi$. Or, if the value of $(\alpha_1 + \alpha_2)$ is assumed to be correct, Eq. 8 can be used as a check on the validity of ϕ, the average flux density.

The values $\alpha_1 = 580$ barns and $\alpha_2 = 105$ barns are rather well established. The value of $\phi = 2.17 \times 10^{14}$ neutrons/cm^2·sec has been tentatively chosen because it gives the best fit to the data based on samples from four test capsules and it is also consistent with values determined by other means. Further data are being obtained for checking the value of ϕ and for ascertaining whether all losses of uranium from irradiated test capsules can be ascribed to burnup.

DETERMINATION OF NICKEL IN HRP FUEL

J. R. Sites H. M. Rosenstock
W. R. Rathkamp

Considerable time has been devoted to the development of a rapid and accurate method of determining small amounts of nickel and large amounts of uranium in the presence of extremely high fission-product activity. The high activity necessitated emphasis on extremely small sample size. The determination of uranium proved to be straightforward, but the nickel determination is still unsatisfactory.

The problem was to vaporize and then to ionize a sufficient quantity of nickel oxide (formed from nickel sulfate). This required a source consisting of a filament coated with nickel oxide and heated to a temperature of about 2000°K. Tungsten was tried and proved to be unsuccessful because of reduction of the nickel oxide by the tungsten and subsequent diffusion of the nickel metal into the body of the filament. Experiments with platinum filaments indicated that this effect was eliminated, as was expected from thermodynamic considerations. However, the maximum filament temperature attainable was too low to yield a sufficient amount of nickel in the vapor phase. With these results at hand, it seems probable that iridium filaments will meet the requirements of chemical inertness and high operating temperature.

Ideally, surface ionization is a much more efficient mode of ion production than electron bombardment and thus permits the use of smaller sample sizes. Experiments with uranium oxide samples of about 1 mg (not subject to the above-mentioned source-filament difficulties) and a tungsten ionizing filament have yielded excellent ion beams suitable for analysis. Nickel, on the other hand, has yielded very poor ion beams by surface ionization. The extent to which the poor yield is affected by the higher ionization potential (7.6 volts for nickel compared with about 4 volts for uranium) is as yet undetermined. This phase of the problem will be investigated further after a better source has been developed, as outlined above.

At present, no conclusions can be drawn as to whether this analytical method can be used with samples small enough to make excessive radiation-hazard protection unnecessary.

ANALYSIS OF OIL SHALE

L. O. Gilpatrick

The Chattanooga strata of oil shale underlying parts of Tennessee have been the subject of considerable interest because of its uranium content. An accurate determination of the uranium values of this mineral has proved to be difficult and elusive because of the complex nature of the material and the low uranium values involved.[4] A suggestion has been made that selected lots of shale could be accurately analyzed for uranium by the method of isotopic dilution.[5] The accuracy to be expected from the isotopic-dilution method should be at least as good as 1% or less of the uranium content.[6]

There has been a request from the AEC that this Laboratory undertake the analyses of a selected group of prepared shale samples by the isotopic-dilution technique. The U.S. Geological Survey has collected and prepared this series of shale samples with great care so that they will be representative and homogeneous, and the Laboratory has received six lots of the material. Considerable effort will be made to obtain the highest possible accuracy in this analysis, and it is anticipated that differences of 1 ppm can be distinguished in the uranium content of these oil shales which is in the range of from 75 to 100 ppm.

Accordingly, three lots of oil shale were secured from J. A. Dean of the University of Tennessee for preliminary study. These materials had been

[4] K. B. Brown, J. M. Schmitt, F. J. Hurst, and D. J. Crouse, *Recovery of Uranium from Oil Shales – Part I. Extraction of Uranium from the Shale Gangue*, Y-564 (Feb. 10, 1950).

[5] Correspondence from K. B. Brown to C. E. Larson, June 27, 1953.

[6] Correspondence from L. O. Gilpatrick to C. R. Baldock, October 14, 1953.

prepared for analysis and had been analyzed by the fluorometric method for uranium by Dean. A procedure which satisfies the conditions for isotopic-dilution analysis of uranium is to ignite slowly about 5 g of ground shale to 750°C and to constant weight. At this point, 1 to 2 mg of uranium enriched in U^{235} is added in accurate amount. The ash is dissolved in mineral acids – mainly hydrofluoric and nitric acids followed by fuming perchloric acid – which do not complex the uranium. The dissolved shale is then treated by the tributyl phosphate extraction method[7] to remove the uranium content, and the uranium is recovered by ammonium hydroxide precipitation and by ignitions to U_3O_8. The oxide is then examined directly in the mass spectrometer by using a high-temperature platinum oven.[8] The isotopic ratio of the U^{235} and the U^{238} is determined by monitoring the UO_2^+ ions.

The results of duplicate determinations of uranium in two of the oil shales are as follows:

Sample		Isotopic-Dilution Analysis (ppm)	Fluorometric Analysis (ppm)
YB-9-E		96.7	92 ± 5
		95.4	
	Average	96.0	
YB-14-E		92.8	66 ± 3
		92.4	
	Average	92.6	

These results represent only the progress to date and are not necessarily the best results to be expected.

NEW ISOTOPIC ABUNDANCES FOR PALLADIUM

J. R. Sites G. Consolazio
Russell Baldock

In the preparation for performing the isotopic analyses on material collected during the recent successful separation of many of the isotopes of the platinum metals group by means of the calutron,[9] the natural abundances for palladium have been redetermined. Some variations have been found from the measurements on palladium reported by Sampson and Bleakney[10] and compiled by Bainbridge and Nier.[11]

The ion source oven used in this investigation was developed by J. R. Walton and was previously described.[12] The essential features are shown in Fig. 2. The center opening is $\frac{1}{8}$ in. long and 0.020 in. in diameter. In this case, a tantalum oven was filled with about 0.5 mg of palladium metal and was heated to various temperatures between 1350 and 2200°C. Ion beams were recorded, and the relative isotopic abundances were calculated. In Table 7 are listed the data of

[7] W. B. Wright, Jr., *Comparison of Tributyl Phosphate and Dibutoxytetraethyleneglycol as Extractants of Uranium*, Y-884 (May 7, 1952).

[8] T. W. Bartlett, *Application of Tributyl Phosphate Extraction of Uranium to Works Laboratory Samples*, K-789 (July 26, 1951).

[9] W. A. Bell, Jr., L. O. Love, C. E. Normand, and W. K. Prater, *Bull. Am. Phys. Soc.* 28, 24 (Sept. 1953).

[10] M. B. Sampson and W. Bleakney, *Phys. Rev.* 50, 732 (1936).

[11] K. T. Bainbridge and A. O. Nier, *Relative Isotopic Abundances of the Elements*, Preliminary Report No. 9, National Research Council (Dec. 1950).

[12] J. R. Walton, *High Temperature Mass Spectrometer Oven*, ORNL-1473 (Jan. 8, 1953).

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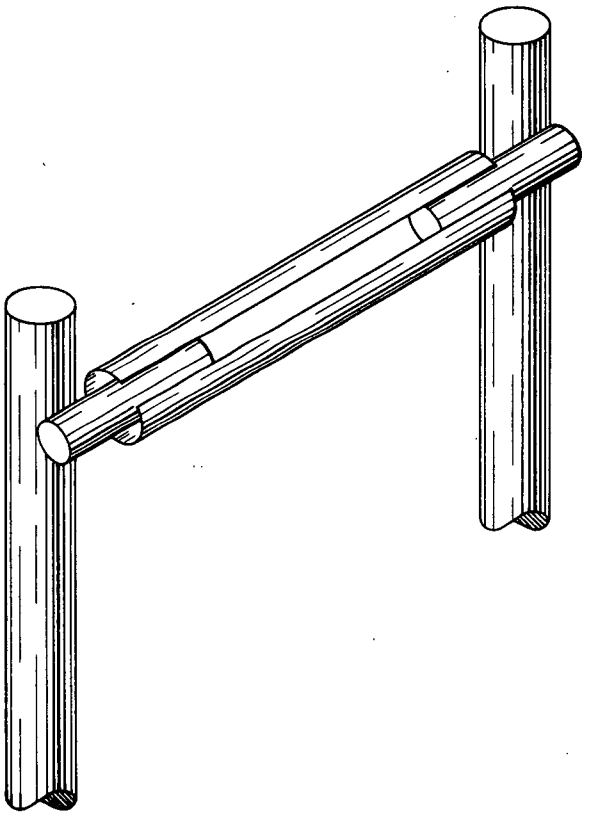

Fig. 2. High-Temperature-Filament Source Oven.

TABLE 7. PALLADIUM ISOTOPIC ABUNDANCE

MASS	DATA OF SAMPSON AND BLEAKNEY (%)	NEW VALUES (%)	CALUTRON SEPARATION AT MASS 108 POSITION (%)
102	0.8	0.96 ± 0.01	0.03 ± 0.01
104	9.3	10.97 ± 0.02	0.20 ± 0.05
105	22.6	22.23 ± 0.03	0.63 ± 0.04
106	27.2	27.33 ± 0.03	2.84 ± 0.04
108	26.8	26.71 ± 0.03	94.19 ± 0.11
110	13.5	11.81 ± 0.03	2.08 ± 0.04

Sampson and Bleakney,[10] the new values reported here, and the isotopic abundances of the palladium collected in the mass 108 position during a calutron separation. The 95% confidence interval was determined for the data presented. Figure 3 shows a bar graph of the new values for the isotopic abundances of natural palladium, with the data of Sampson and Bleakney indicated by cross bars. Figure 4 shows typical scans of natural palladium and of palladium enriched in the mass 108 isotope.

By use of the mass defects of palladium reported by Halsted[13] and the new isotopic abundances reported here, a molecular weight of 106.5 ± 0.1 is obtained, as compared with 106.6 determined by Sampson and Bleakney[10] and with 106.4 to 106.9 reported from chemical determinations.[14,15,16]

[13] R. E. Halsted, *Phys. Rev.* 88, 666 (1952).

[14] G. P. Baxter, O. Hönigschmid, and P. LeBeau, *J. Am. Chem. Soc.* 58, 541 (1936).

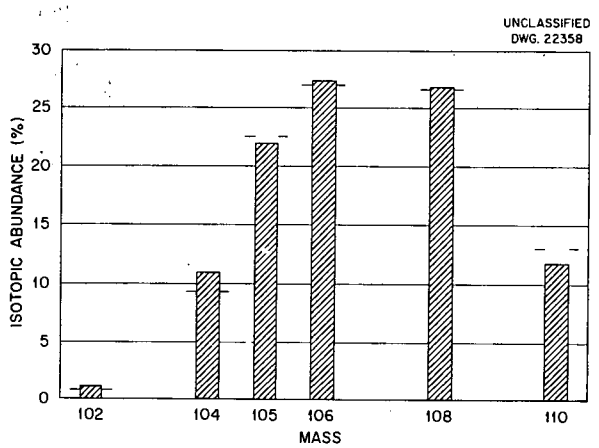

Fig. 3. Isotopic Abundances of Natural Palladium.

CONSTITUTION OF METAL HALIDE VAPORS

H. M. Rosenstock J. R. Sites

A consideration of the constitution of metal halide vapors was initiated with the observation that the mass spectra of alkali halide vapors indicated the presence of appreciable amounts of ions such as Na_2Cl^+, K_2Cl^+, and Li_2Cl^+. Only a few cases of polymerization have been discussed in the literature;[17] outstanding among them is the work of Brewer and Lofgren[18] on the solid-vapor equilibriums of the cuprous halides. The marked curvature in the customary log p vs. $1/T$ plots showed the presence of more than one vapor species. Brewer and Lofgren concluded that the two species were CuCl and Cu_3Cl_3 in the case of cuprous chloride. Some evidence for the existence of Cu_3Cl_3 has been obtained here by means of mass spectrometer studies.

The existence of the alkali halide polymers may require the reinterpretation of many reports dealing with thermodynamic studies on these compounds.

EQUIPMENT AND TECHNIQUES

New Mass Spectrometer Tube (MSB-2)

G. F. Wells

The recently constructed all-metal mass spectrometer tube and the radiation-shielded housing

[15] G. I. Kemmerer, *J. Am. Chem. Soc.* 30, 1701 (1908).

[16] O. L. Shinn, *J. Am. Chem. Soc.* 34, 1448 (1912).

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

[18] L. Brewer and N. L. Lofgren, *J. Am. Chem. Soc.* 72, 3038 (1950).

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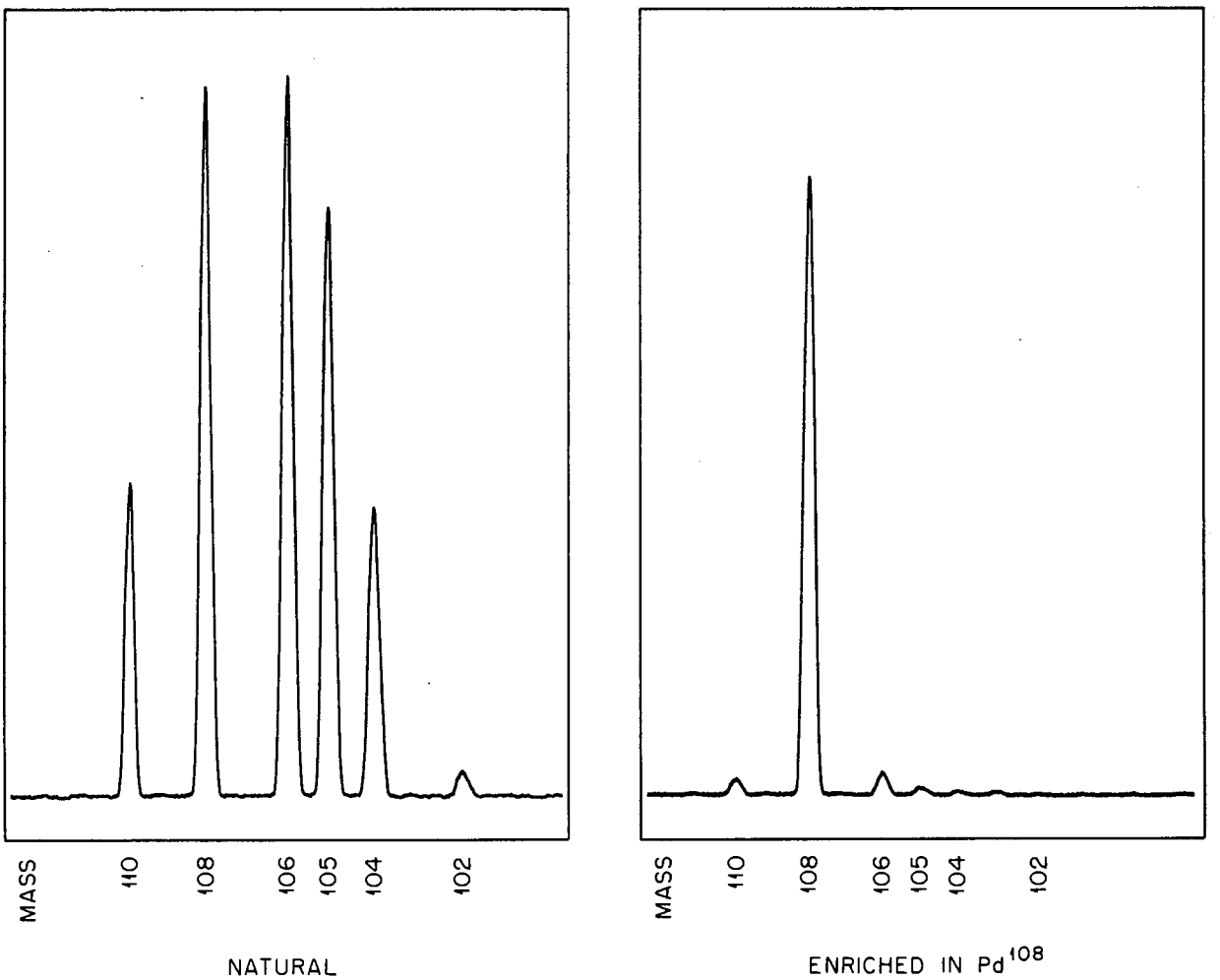

Fig. 4. Typical Scans of Natural and of Enriched Palladium.

have been assembled and checked for alignment. By using a standard high-temperature source and a vibrating-reed detector at the receiver, several peaks were observed in the low-mass range from residual gases present. High voltage, emission regulation, and magnet supply were obtained from MSB-1, the G-E mass spectrometer.

Filament Power Supplies

W. R. Rathkamp

A high-current, stabilized, a-c power supply for sample filaments was designed, and two were constructed and installed. The supplies are regulated to greatly minimize current fluctuations due to line-voltage changes or load variations. There is also a provision for scanning data by increasing the filament current at a slow, controllable rate. This will permit continuous studies of ion current vs. sample temperature.

Determination of Small Leaks

J. R. Walton

A number of metal floats were received and were leak tested with the aid of a mass spectrometer. The maximum diameter of the floats was 5 in., and a stem 20 in. long was welded to one end. These float assemblies were welded closed in a dry box under a pressure of helium slightly greater than

atmospheric. A suitable container into which the floats could be placed was constructed, and the container was evacuated. Monitoring of the helium peak was used to detect leakage from the float.

The evacuation chamber consisted of a heavy-walled (6 in. by 36 in.) pyrex pipe with cast-iron flanges to hold ½-in. brass end plates. Teflon "O" rings were used as gaskets because of their desirable low-outgassing qualities. One end of the container was connected to a Welch Duo Seal pump and to a McLeod gage, and the container was then evacuated to 1 micron. To the other end plate, a suitable fitting was attached which was used to connect to the mass spectrometer sample system. Thus, a sensitive method was provided for detecting extremely small leaks that might exist in the floats.

PUBLICATIONS

Paper presented:

1. J. R. Sites, G. Consolazio,[19] and Russell Baldock, "Isotopic Abundances of Palladium," presented at the American Physical Society, Albuquerque, New Mexico, September 2-5, 1953.

[19] Watertown Arsenal, Watertown, Massachusetts.

RESEARCH AND DEVELOPMENT STABLE ISOTOPES

P. S. Baker F. R. Duncan
H. B. Greene

HILSCH TUBE

P. S. Baker W. R. Rathkamp[1]

Investigation of the Ranque-Hilsch (vortex) tube has been terminated for the time being, and the completed report is being processed for publication.

The additional studies performed during this report period were aimed at an attempt to establish a theory and, in a few instances, were for checking previous data; the studies have supported earlier conclusions. Although a complete theory is still lacking, the evidence points to a tube operation whose effects are dependent upon the combination of an adiabatic expansion of a portion of the inlet gas, a "viscous-shear" effect which transfers energy from the center of the tube to the outside layers of gas, and an energy release associated with the turn-around of gas molecules at the stagnation point of the tube.

Two additional reports which have been received[2,3] mention that analyses of the outlet gases from the vortex tube show no measurable separation of the gas constituents.

Work with a Uniflow tube (Fig. 5), which is designed to show the effects of centrifugation, showed results that were comparable with those obtained with the regular vortex tube:

	N_2^{28}-to-O_2^{32} Ratio	N_2^{28}-to-N_2^{29} Ratio
Takeoff (periphery)	4.49 ± 0.02	134 ± 1
Inlet	4.53 ± 0.03	136 ± 2

The averages of these particular samples showed deviations which suggested centrifugation, but the values cannot at this time be considered significant because they are still within the limits of experimental error.

Samples of ordinary air taken from the room, placed in sample bottles, and submitted for mass analyses as controls showed results which could very well have come from the regular vortex tube – results which show definite "enhancement" (Table 8) when the data are arbitrarily arranged.

METAL REDUCTION AND RELATED WORK

F. R. Duncan H. B. Greene
P. S. Baker

Activities during this report period have continued to be those related to calutron operations and to the research and development projects associated with other isotope-separation methods.

[1] Mass Spectrometer Laboratory.

[2] A. F. Johnson, *Can. J. Research* 25(F), 299 (1947).

[3] J. E. Corr, *The Vortex Tube*, GE Data Folder No. 45289 (July 1948).

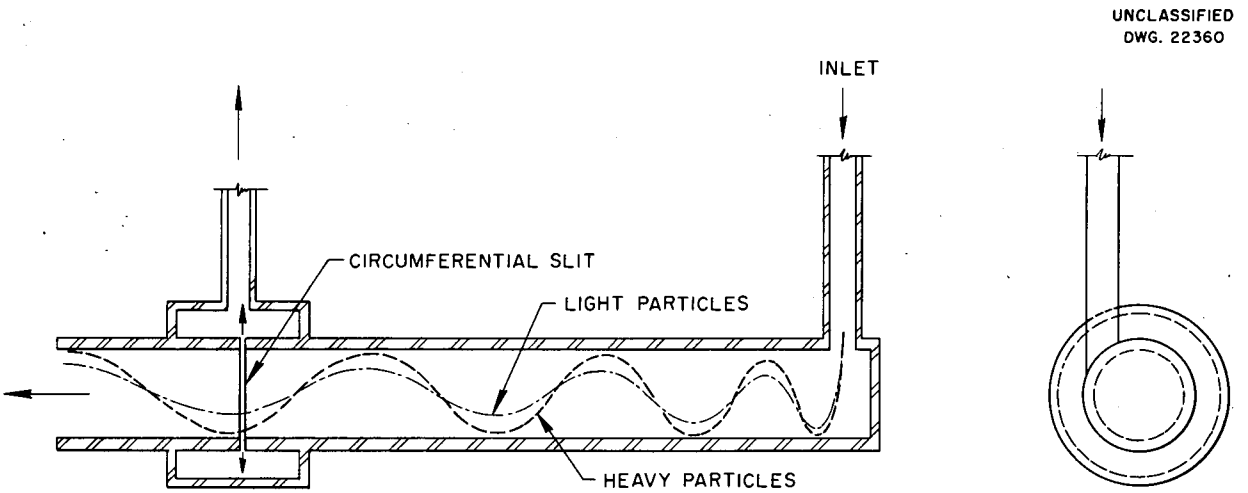

Fig. 5. Uniflow Vortex Tube.

TABLE 8. CONTROL SAMPLES OF AIR

SAMPLE*	N_2^{28}-TO-O_2^{32} RATIO
1a	5.22
1b	5.17
1c	5.17
Average	5.19 ± 0.02
2a	5.27
2b	5.27
2c	5.23
Average	5.26 ± 0.02
3a	5.30
3b	5.23
3c	5.20
Average	5.24 ± 0.03

*Each sample was subjected to three separate scans (*a*, *b*, and *c*) on the mass spectrometer.

1. A method for the fusion of finely divided rare-earth chlorides (prepared by the Isotope Chemistry Group) into a solid form has been developed and applied to calutron charge material.

2. Reduction of Li^6 metal from its chloride by the barium-reduction process already described has continued.[4] The barium metal is now vacuum distilled prior to use and produces a better grade of lithium metal as end product.

3. Development of equipment and of a safe procedure for preparing small experimental quantities of Li^6H was undertaken, with the preliminary work being done with the normal metal. The compound formed readily at 800°C in an atmosphere of hydrogen. Preparation of the hydride from enriched isotopic metal is awaiting availability of the metal, which at present is not obtainable owing to depletion of the inventory by allocation to other experimental projects.

4. A technique was developed for dissolving lithium metal in mercury and then determining the amount of dissolved material by titration with acid. This technique is to be applied to the measurement of the solubilities of the individual lithium isotopes.

5. As a direct result of the observations by O. C. Yonts and by the Calutron Operations Group that gadolinium is responsible for high electron emission, several grams of metal were prepared from the chloride by reduction with calcium metal[5] at a temperature of 1350°C. (Neodymium metal had been prepared previously by a similar process at a lower temperature, but it was necessary to redesign the equipment for the gadolinium reduction.) Part of the metal prepared in this way was used by Yonts to measure its electron emissivity. The remainder was used in connection with calutron filaments –

[4] P. S. Baker, F. R. Duncan, and H. B. Greene, *Lithium Metal Production*, ORNL CF-53-4-185 (May 1, 1953).

[5] F. H. Spedding and A. H. Daane, *J. Am. Chem. Soc.* 74, 2783 (1952).

first, to try to prepare pure gadolinium filaments and then, since these were unsatisfactory, to modify conventional tantalum filaments by embedding pieces of gadolinium metal in them and heating until the filaments were coated with the gadolinium.

6. Preliminary procedures dealing with the adsorption (or absorption) of inert gas ions in metal targets have been extensively investigated. The purpose of this program is to obtain data relevant to the design of collectors for the calutron collection of inert-gas isotopes. Although the program is still in its infancy, some tentative observations can be presented:

a. Targets struck by 3-kev ions in the vapor-pressure magnet equipment exhibited virtually no adsorption, whereas targets bombarded at 30 to 40 kev in the calutrons have yielded gas quantities in close agreement with those observed at 60 kev and reported by Koch.[6]

b. Targets outgassed at 750°C in vacuum prior to exposure to calutron beams gave considerably better results than those not outgassed in this manner. The time for complete outgassing of the target was found to be several hours. The outgassed targets exhibited a partial readsorption of atmospheric gases when they were re-exposed to the air, but the extent of readsorption was small when compared with the amount present before outgassing. (The amount readsorbed, however, will be of considerable importance when an attempt is made to obtain collected isotopes of high purity.)

c. Targets, folded (plaited) so that the projected exposed area was less than the true area, adsorbed larger volumes of gas ions than the flat targets, suggesting that oblique surfaces can be used to increase collections.

d. Targets of a variety of different metals were bombarded, and the quantity of gas adsorbed by each was determined. Argon was used as the charge material (see Table 9). The data represent single calutron runs for each metal; however, the runs are to be repeated and more metals are to be tried. It is of interest to note that Koch's predicted saturation value for silver is 0.014 cc/in.^{2}. As yet, no effort has been made to determine the significance of target thickness.

e. An apparatus was built for the collection of the adsorbed gases as they are outgassed from the targets. It is shown in Fig. 6 and its operation is tantamount to that of a hand-operated Toepler pump.

TABLE 9. ARGON ADSORPTION ON METAL TARGETS

METAL TARGET	ADSORPTION OF ARGON (cc/in.^{2})*
Silver	0.013
Copper	0.014
Aluminum	0.029
Nickel	0.032
Brass	0.015
Monel	0.003
Hastelloy	0.015

*All quantities were reduced to standard conditions.

Fig. 6. Apparatus for Collection of Adsorbed Gases.

[6] J. Koch, *Nature* 161, 566 (1948).

The preparation of an unclassified, calutron ion source and an isotope collector for use at the American Museum of Atomic Energy was completed and they are now on display at the Museum.

PUBLICATIONS

Papers accepted for presentation or publication:

1. P. S. Baker, F. R. Duncan, and H. B. Greene, "Thermochemical Reduction of Lithium," to be presented at American Association for the Advancement of Science, Boston, December 27, 1953.
2. P. S. Baker, F. R. Duncan, and H. B. Greene, "Preparation of High-Purity Lithium Metal," to be published in *Science*.

STABLE ISOTOPE SEPARATION RESEARCH AND CALUTRON OPERATIONS

C. E. Normand, Department Head

INTRODUCTION

Eight elements (platinum, ruthenium, neodymium, gadolinium, iridium, samarium, potassium, and lanthanum) were processed in the calutrons during the period from June through November 1953. Gadolinium, platinum, ruthenium, and iridium were processed for the first time and required the development of a source unit capable of operating at temperatures up to an estimated 2800°C. All collections were made by operating personnel under the direction of W. A. Bell, R. L. Caldwell, G. J. Fisher, C. V. Ketron, L. O. Love, and K. A. Spainhour. Equipment maintenance was under the direction of W. K. Prater, and G. M. Banic was in charge of electrical maintenance.

COLLECTIONS

Platinum, Series GH-XBX

Collection of the isotopes of platinum (Pt^{190}, Pt^{192}, Pt^{194}, Pt^{195}, Pt^{196}, Pt^{198}) was started on March 24 and completed on June 18, 1953. The bombardment-heated source units and the operating procedures remained essentially as reported previously,[1] except for improvements in source design as noted in a later section of this report.

Of the 160 runs comprising the series, 81% attained innage for an average run time of 8.5 hr and an average production rate of 3.15 ma. Of the last 32 runs of the series, however, 97% attained innage for an average of 13.2 hr, and the average production rate was 4.18 ma. The following estimated weights, in grams, of platinum isotopes were collected:

Pt^{190}	0.26
Pt^{192}	0.30
Pt^{194}	12.78
Pt^{195}	4.30
Pt^{196}	11.36
Pt^{198}	0.73
Pt^{200}*	0.25
Total	29.98

Ruthenium, Series GJ-XBX

Collection of the seven isotopes of ruthenium (Ru^{96}, Ru^{98}, Ru^{99}, Ru^{100}, Ru^{101}, Ru^{102}, Ru^{104}) was started on June 18 and completed on August 18, 1953.

The metal was vaporized in an electron-bombardment-heated source of the type in use at the conclusion of the platinum series. The estimated operating temperature for ruthenium was 2600°C, as compared with 2150°C for platinum. At this higher temperature, both service life and production rate were lower than for platinum, and high drains, attributed to vaporization of graphite source parts, became noticeable.

Innage was attained in 89 of the 93 runs comprising the series, and the average length of all innage runs was 9.1 hours. The production rate averaged 2.45 ma and was limited at most times by the high drains resulting from graphite vapors.

[1] *Stable Isotope Research and Production Semiann. Prog. Rep. May 31, 1953*, ORNL-1563, p. 22.

*Collector pocket was placed in this position although the isotope is not known to exist.

The following estimated weights, in grams, of ruthenium isotopes were collected:

Ru^{96}	0.42
Ru^{98}	0.23
Ru^{99}	0.74
Ru^{100}	0.44
Ru^{101}	1.08
Ru^{102}	3.00
Ru^{104}	1.61
Total	7.52

Iridium, Series GL-XBX

Collection of the isotopes of iridium (Ir^{191}, Ir^{193}) was started on August 11 and completed on October 7, 1953.

The metal was vaporized in an electron-bombardment-heated source at an estimated temperature of 2800°C. The design of the source block, disposition of the bombarding electron beams, and extent of heat shielding required for attaining this temperature are discussed in a later section of this report (cf. "Research and Development"). Low ion output indicated that even higher temperatures would be beneficial; however, excessive vaporization of the graphite source block, as manifested by high drains and the appearance of carbon deposits on adjacent cooler surfaces, made higher operating temperatures impractical.

The series consisted of 115 runs, 97 of which attained innage for an average of 9.7 hr and an average production rate of 0.51 ma. The following estimated weights, in grams, of the two iridium isotopes were collected:

Ir^{191}	1.34
Ir^{193}	2.12
Total	3.46

Neodymium, Series GI-XAX

Collection of neodymium isotopes (Nd^{142}, Nd^{143}, Nd^{144}, Nd^{145}, Nd^{146}, Nd^{148}, Nd^{150}) was started on April 16 and completed on June 25, 1953. Equipment and operating characteristics remained essentially as reported previously.[1]

The series consisted of 46 runs, 42 of which produced innage. Almost half the innage runs had an average life of 50 hr, and the average production rate for the series was 11.38 ma. The estimated weights, in grams, of neodymium isotopes collected are as follows:

Nd^{142}	18.32
Nd^{143}	9.24
Nd^{144}	14.93
Nd^{145}	6.01
Nd^{146}	8.28
Nd^{148}	2.38
Nd^{150}	1.75
Total	60.91

Gadolinium, Series GK-XAX

Gadolinium was processed in the calutrons for the first time during the period from June 25 to August 24, 1953. All the gadolinium isotopes (Gd^{152}, Gd^{154}, Gd^{155}, Gd^{156}, Gd^{157}, Gd^{158}, Gd^{160}) were collected, and, in some runs, additional pockets were placed in the receiver to collect the ions falling in neighboring mass positions.

A medium-high-temperature source, operating at about 800°C with a $GdCl_3$ charge, was used. A $^3/_{16}$-in., zero-degree, ion exit slit, standard zero-degree electrodes, a 170-mil tantalum filament, and a carbon charge bottle were initially used. Seepage of charge through the walls of the carbon bottle made it necessary to replace this bottle with one made of stainless steel, and the later addition of laminated, carbon heat shielding to the front of the arc chamber modified the ion exit slit from a zero- to a 30-deg type. Ion exit slits of different widths and accelerating electrodes of different configurations were tried, but these changes resulted in no improvement in performance and were not retained.

In general, calutron operation during the collection of gadolinium followed the pattern set by the rare earths previously processed and was characterized by recurrent sparking flurries, relatively low output, poor control of focus, and the formation of deposits on the electrodes and inside the arc chamber. Deposits inside the arc chamber were removed by running the chamber at a higher temperature, but the other difficulties persisted throughout the collection series.

Pronounced activation of the tantalum filament was a disturbing effect in that cycles of extreme activation and deactivation, accompanied by wide

variations in filament current and arc conditions, occurred periodically and almost uncontrollably.

The isotopes were received in graphite pockets placed behind ⅛-in. defining slits. Because of space limitations, no internal shielding was used, and pockets 155, 156, and 157 were not electrically insulated so as to permit individual monitoring. In some of the later runs, pockets were placed to collect ions falling in the 159, 161, 162, 163, and 164 positions.

The series consisted of 37 runs, 32 of which attained innage for an average of 40.7 hours. Although a peak output of 28 ma was obtained, the average production rate during innage was only 8.3 ma. The following weights, in grams, of gadolinium isotopes were collected:

Gd^{150}*	0.14
Gd^{152}	0.42
Gd^{154}	2.98
Gd^{155}	11.07
Gd^{156}	15.37
Gd^{157}	11.99
Gd^{158}	10.34
Gd^{160}	11.02
Total	63.33

Samarium, Series GM-XAX

Separation of the isotopes of samarium (Sm^{144}, Sm^{147}, Sm^{148}, Sm^{149}, Sm^{150}, Sm^{152}, Sm^{154}) was started on August 20 and completed on October 20, 1953.

A fused $SmCl_3$ charge was run in a medium-high-temperature source at 750 to 850°C. Because of the breakage of carbon charge bottles when they are reused, either stainless steel or new carbon bottles were used. Initially, standard zero-degree slit and electrode geometry was used, but this was later changed to 30-deg geometry to permit the installation of heat shielding over the front of the arc chamber. Collections were made in graphite pockets placed behind ⅛-in. defining slits and separated by mica where closed clearances occurred.

Calutron operation during samarium collection was probably the smoothest encountered in rare-earth separations. Sparking occurred infrequently, and there was a minimum of filament activation; however, beam focus was erratic and consistently poor, although it was somewhat improved by operation at reduced output.

Of the 39 runs comprising the series, 34 attained innage for an average of 38.8 hr per run and an average production rate of 10.94 ma. The following estimated weights, in grams, of samarium isotopes were collected:

Sm^{142}*	0.21
Sm^{144}	1.55
Sm^{147}	10.54
Sm^{148}	9.15
Sm^{149}	10.19
Sm^{150}	6.60
Sm^{152}	24.94
Sm^{154}	17.99
Total	81.17

Potassium, Series GN-XBX

A special collection of potassium isotopes (K^{39}, K^{40}, K^{41}) was started on October 7 and is still in progress. The aim of this collection is to produce K^{40} of the highest possible purity from charge material in which the K^{40} content has been increased from 0.012 to 0.03% by reactor irradiation. The value of this material requires that it be recovered, and because of the limited amount available, recycling may be necessary. To minimize the problems and to ensure maximum separation throughout the processing of the enhanced material, a number of preliminary runs were made with natural potassium iodide charge. During the course of these runs, both equipment and operating techniques were perfected before the high-purity collections were undertaken.

On the basis of the preliminary runs, the best equipment for the high-purity collections appeared to be a medium-temperature source with supplementary heat shielding, either a standard or deep-floating-anode arc chamber, a ⅛-in., ion exit slit, and zero-degree electrodes of two-thirds standard size. At the receiver, copper pockets are placed behind ⅛-in. defining slits. Water cooling has been extended to the bottom surface of the K^{39} pocket; the K^{40} pocket is surrounded by carbon shielding and is coated with clear Glyptal to insulate the pocket from the shielding.

*Collector pocket was placed in this position although the isotope is not known to exist.

During operation, steady drains were not severe, but sparking was prevalent. There were no filament failures, and several runs went to scheduled terminations; however, severe erosion of the K^{39} pocket may well be the chief factor that limits run time. Focus was good at moderate output (10 to 15 ma) but was poor at high output (47 ma). Thus, the obtaining of high purity will require some sacrifice of output.

At the conclusion of the preliminary runs (November 17), the average run time was 87.4 hr, and the average output was 11.3 ma. Separation effectiveness, as indicated by the ratio of the monitored K^{39} to K^{40} ion peaks, varied from 736 to 3465, with an average of 1932; and K^{40} collected at a ratio of 1785 assayed 5.7%. In the later runs, ratios of 2500 or better were commonly attained. During the preliminary runs, the following amounts, in grams, of potassium isotopes were collected:

K^{39}	17.6424
K^{40}	0.0081
K^{41}	1.0756
Total	18.7261

Lanthanum, Series GO-XAX

Collection of lanthanum isotopes (La^{138}, La^{139}) was started on October 20 and is still in progress. Substantial enrichment of the rare La^{138} isotope is the chief objective of this series. Small separation at the receiver (0.366 in.) and the fact that the minor constituent (La^{138}) lies on the broad, low-energy side of the major constituent (La^{139}) are conditions unfavorable to good separation. Moreover, in previous separations of lanthanum, focus was consistently poor, and the La^{138} abundance was increased from 0.089 to only 0.597%. In the present series, focus has been somewhat improved, although it is still not good.

A medium-high-temperature source is used with a $\frac{3}{16}$- or a $\frac{1}{8}$-in., 30-deg, ion exit slit and 30-deg electrodes. Operation with a $LaCl_3$ charge is at approximately 650°C.

The ions are received in purified carbon pockets placed behind $\frac{1}{8}$-in. defining slits and enclosed in a single carbon shield. The front of the 139 pocket is cut at a 45-deg angle and is spaced $\frac{1}{16}$ in. behind the receiver faceplate.

Nitrogen support gas is used at all times, and moderate sparking and some filament activation are encountered. Material collecting on the arc front forms wide bridges across the ion exit slit, thereby reducing output and disturbing focus.

The series to date (November 13) comprises 15 runs of 37.2 hr average length and 7.1 ma average output. Estimated collections, in grams, of the lanthanum isotopes are

La^{138}	0.6028
La^{139}	17.1688
Total	17.7716

RESEARCH AND DEVELOPMENT

The electron-bombardment-heated source used in the separation of palladium and described in a previous report[1] was further developed during the present report period. Development was directed primarily toward attaining the higher operating temperatures required for processing platinum, ruthenium, and iridium. By increasing the amount and improving the quality of heat shielding around the source block, by reducing the size of the source block, arc chamber, and charge cavity, and by improving the disposition of the bombarding electron beams on the source block, temperatures ranging up to an estimated 2800°C were obtained. The above-mentioned elements were processed in this temperature range, but operating experience indicates that excessive vaporization of the graphite source parts precludes operation at significantly higher temperatures.

Significant features of the bombardment-heated source in its final form are shown in Fig. 7. Two views of the assembled source show the arrangements of arc filament, bombardment filament, and bombardment grid, and also show the heat shielding of the source front.

Improvements in source-block design and in the disposition of bombarding electron beams are shown in Fig. 8. In these views, the source blocks have been sectionalized to show the erosion resulting from electron bombardment. The upper picture shows a block of the single-cavity type used in early models of the unit. The charge was distributed along the trough at the bottom of the arc chamber, and bombardment was by a single, broad beam of electrons. The center picture is of a block of the same type which has been bombarded by four electron beams. The lower picture shows a block of the type finally developed. A separate chamber is provided for the charge, and the

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PHOTO 20626

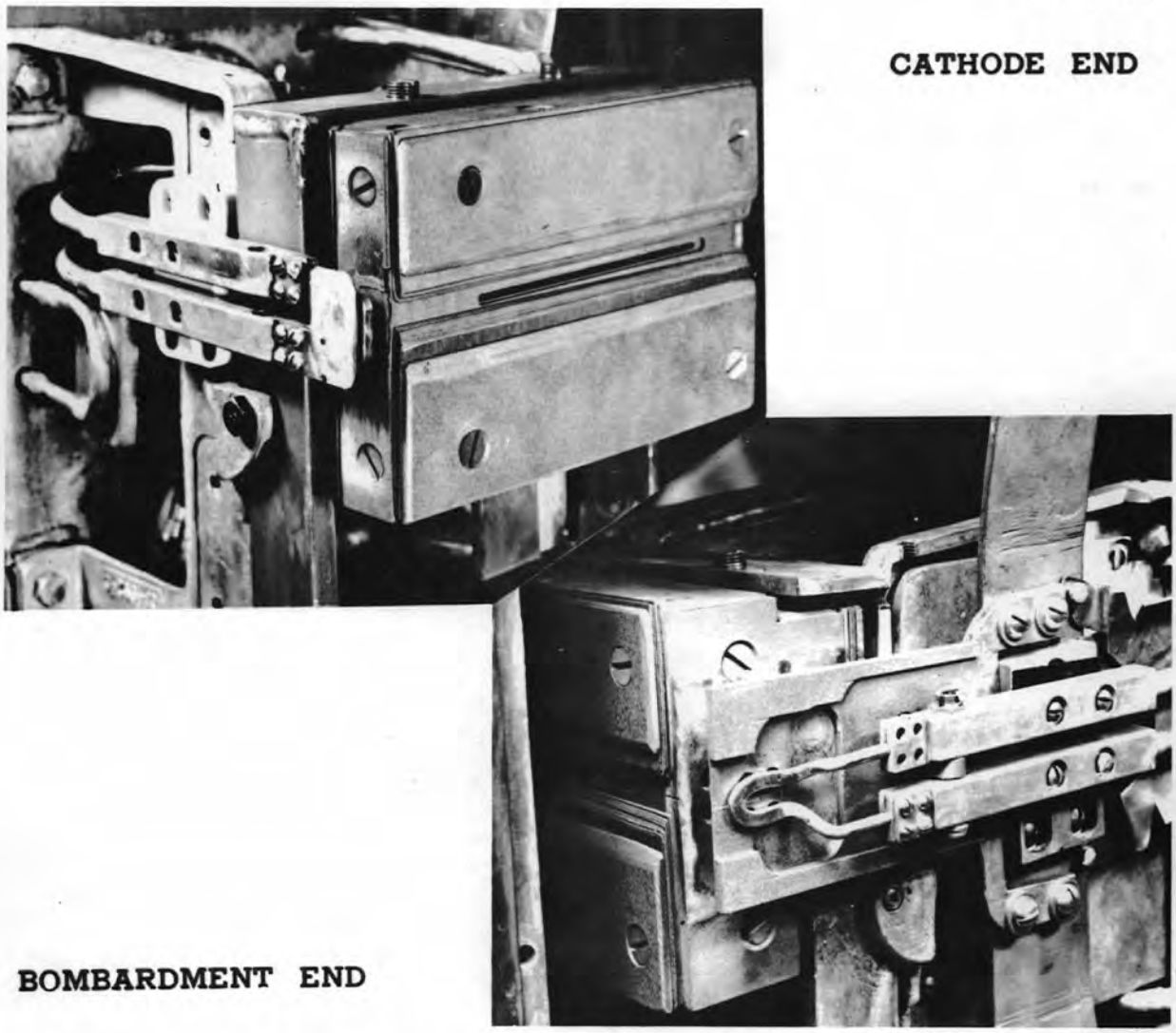

Fig. 7. Final Assembly of Bombardment-Heated Source.

bombarding electron beams are equally spaced around this cavity.

The evolution of heat shielding is illustrated in Fig. 9. The only heat shielding in the palladium source is that provided by thin graphite sheets surrounding the source block and covering the graphite support structure. In the ruthenium source, the source block was surrounded by fine carbon dust to a thickness of approximately 1 in., and the graphite support structure was lined on the inside and covered on the outside with a $\frac{3}{8}$-in. thickness of thin carbon sheets.

With these improvements in source design and heat shielding, the operating temperature of the unit was increased from the 1750°C required for palladium to the 2800°C at which iridium was processed.

The observance of filament activation during the processing of gadolinium led to a number of experimental runs being made in which the filaments were coated or inlaid with gadolinium. Generally, these filaments showed evidence of activation during early stages of operation but reverted to essentially normal emission after a short time.

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PHOTO 20627

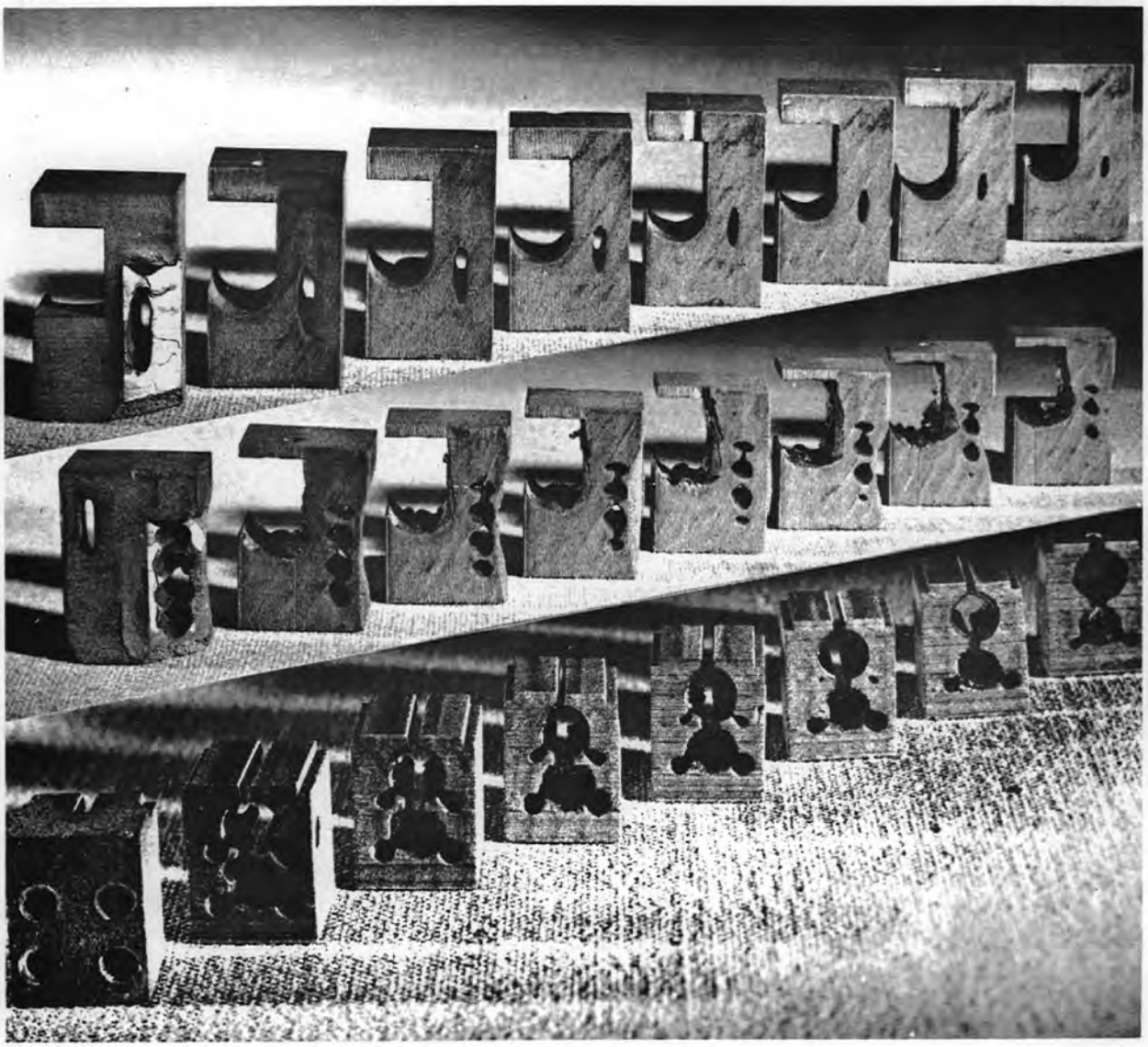

Fig. 8. Sectionalized Source Blocks.

This behavior is in general agreement with previous observations on other activating agents and further demonstrates the inability of activation coatings to stand up under calutron filament conditions.

As an approach to the general problem of processing the inert gases, several exploratory runs were made in which argon was used as the test gas and the ions were collected by allowing them to impinge on metal targets. Choice and pretreatment of target material and the development of product-recovery techniques are essential parts of the over-all problem. Results to date are largely qualitative and far too fragmentary to indicate more than the fact that impinging ions are retained in, and eventually recovered from, the targets.

Filament-life experiments by O. C. Yonts of the Isotope Analysis Methods Department were continued during the first three months of this period, utilizing 431 hr of calutron time. Titanium was run in these experiments; however, no collections were made, and the results are given in the section

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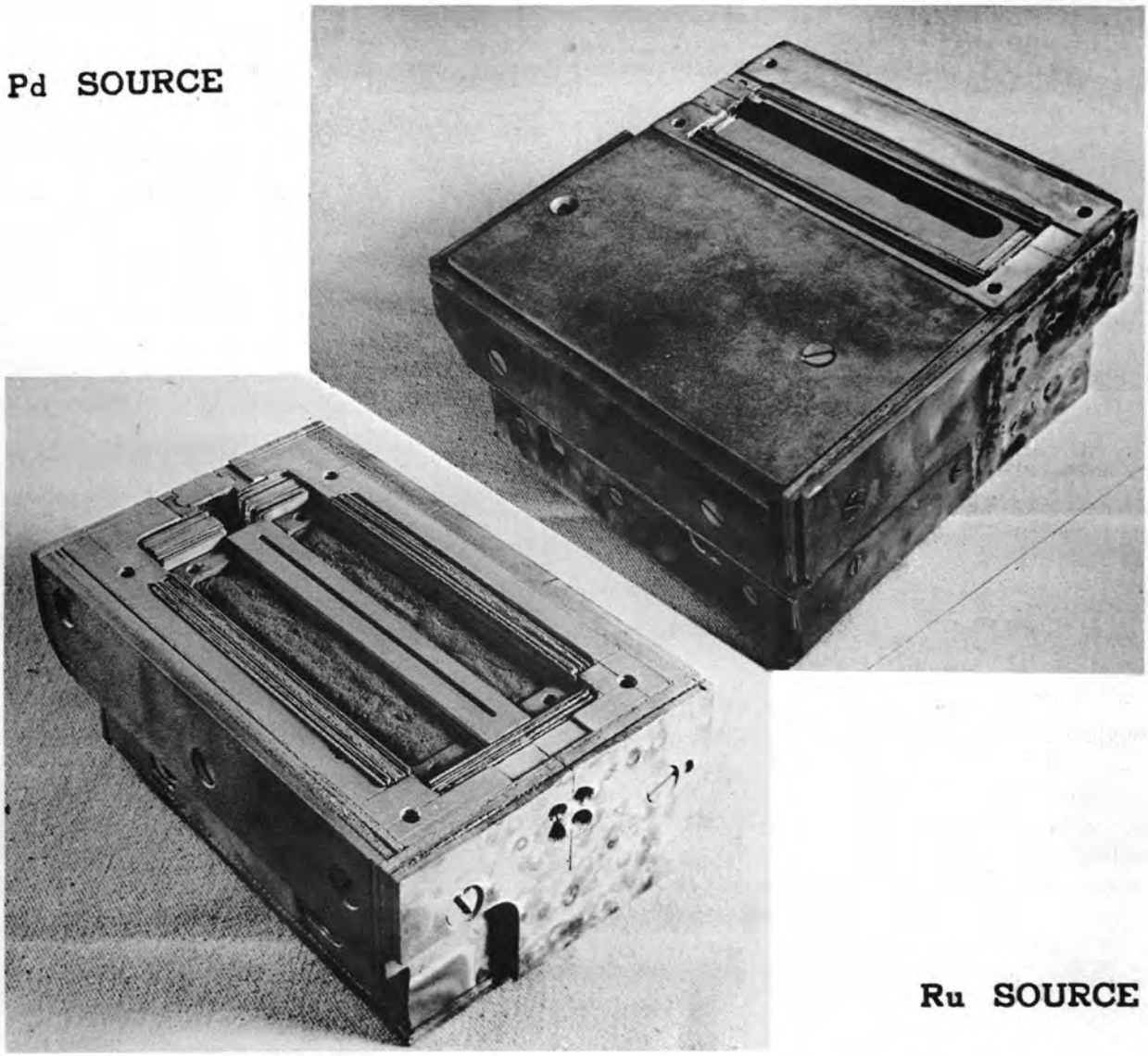

Fig. 9. Source-Block Heat Shielding.

of this report covering the activities of the Isotope Analysis Methods Department.

PUBLICATIONS

Reports issued and papers presented:

1. C. E. Normand, *Ionic Centrifuge Method of Separating Isotopes of Uranium*, ORNL CF-53-7-220 (July 1953), an evaluation report to the Atomic Energy Commission.
2. W. A. Bell, Jr., L. O. Love, C. E. Normand, and W. K. Prater, "Electromagnetic Separation of Pd, Pt and Ru Isotopes," paper presented at the Albuquerque meeting of the American Physical Society, September 2-5, 1953.

STABLE ISOTOPE CHEMISTRY, RESEARCH, AND PRODUCTION

Boyd Weaver, Department Head

INTRODUCTION

The Stable Isotope Chemistry, Research, and Production Department continued to prepare charge materials for the calutrons, recover and purify separated isotopes for the inventory, and prepare for shipments such quantities of the separated isotopes as have been requested for use in research programs both within and outside the jurisdiction of the AEC. Activities during this period have included the development of quantitative chemical procedures for an unusually large number of new elements and the recovering and refining of several expensive and rare materials. The supplying of charge materials has involved the development and application of a new process for the separation of the rare-earth elements.

PREPARATION OF CHARGE MATERIALS

C. W. Sheridan C. F. Harrison
W. C. Davis

Since calutron operations during this period have been confined almost entirely to the palladium, platinum, and rare-earth groups, chemical operations involving these elements were carried on throughout the period. The use of the elemental form of the palladium-platinum metals required no laboratory preparation of the primary charge materials. However, the recovery of these expensive materials from various calutron parts, to conserve these difficultly obtained elements, was more laborious than most charge preparations. The rare earths required the continual recovery and reprocessing of material from calutron parts and the preparation of anhydrous chlorides. The production of the individual elements is discussed separately from charge-material preparation, although the preparation is the primary reason for the rare-earth chemical separation activities.

Palladium-Platinum Group

Impure palladium, platinum, and iridium are recovered from calutron carbon parts by ignition in a muffle furnace with an oxygen atmosphere. Ruthenium must be ignited in a quartz tube fitted with a trap for the collection of volatile oxides. Most of the ruthenium deposits as RuO_2 in the cooler parts of the tube.

Palladium metal is dissolved in nitric acid, and the addition of ammonium chloride causes quantitative precipitation of ammonium chloropalladate $(NH_4)_2PdCl_6$, provided that the palladium is first oxidized in solution with chlorine. Small quantities may be more conveniently precipitated by means of dimethylglyoxime. Both precipitates reduce to palladium metal when they are ignited. Thus far, metal produced by either method has required reprocessing through this cycle.

Platinum metal dissolves only slowly, even in aqua regia. It may be precipitated as ammonium chloroplatinate only after all the nitrate has been removed by repeated treatment with hydrochloric acid. The metal is then formed by ignition.

Iridium resists all liquid solvents. It can be converted to a water-soluble form by chlorination at high temperatures in quartz apparatus when it is mixed with sodium or potassium chloride. The development of this process is still in progress. A process is also under development for the reduction of ruthenium oxide to the metal.

The recovery of these elements from metal calutron parts presents special difficulties. Palladium can be dissolved along with copper or from stainless steel. Platinum and iridium will be left out of solution when copper is dissolved in nitric acid or when stainless steel is dissolved in hydrochloric acid. The recovery from large calutron parts is cumbersome and inefficient.

Rare Earths – Neodymium, Samarium, and Gadolinium

Anhydrous chlorides are made by heating the oxides with an excess of ammonium chloride and then removing the last of the ammonium salt by sublimation from a tube furnace at reduced pressure. Rare earths recovered from calutron parts by washing with nitric acid are purified by repeated precipitations with ammonium hydroxide and oxalic acid used alternately. Several kilograms of oxides were conserved for re-use.

CHEMICAL REFINEMENT

R. L. Bailey C. F. Harrison
W. C. Davis L. Royer[1]

All the isotopes of palladium and platinum were added to the inventory for the first time; and refinement was completed on the first production of gadolinium isotopes, except for the removal of other rare earths. New series of neodymium, boron, sulfur, silicon, and germanium were refined. Progress was made in the development of methods for the refinement of ruthenium and iridium, and the refinement of a new series of titanium isotopes was nearly completed. Some details of production methods and results follow. The lowering of the limits of spectrographic detection and estimation has increased the number of impurities observed in refined isotopes.

Neodymium

A rather simple procedure has been standardized for the preliminary purification of rare-earth isotopes collected in carbon pockets. The pockets are burned, and the ash is dissolved in hydrochloric acid. Separation from copper and calcium is made by precipitation of the rare earths with ammonium hydroxide; this may be repeated once or twice if the copper persists. The hydroxides are redissolved in hydrochloric acid, and the rare earths are precipitated by oxalic acid. This step also requires repetition if the iron content is high. The oxides formed by ignition of the oxalate are usually free of all elements except rare earths.

Rare-earth impurities can be removed quantitatively only by elution with a buffered citric acid–ammonium citrate solution from a strongly acidic, cation-exchange resin. Nearly all the treatment of isotopes has been in a 1¼-in.-dia column containing about 1 meter of Dowex-50 finer than 250 mesh. With a head of 8 ft on the eluting solution, a flow rate of about 3 ml/min has been maintained. The time required for elution and the degree of separation attained depend on the quantity of material processed, but with quantities of less than 1 g of oxide, there is virtually complete separation between neodymium and samarium. The effluent is collected in fractions, and the rare-earth content is precipitated by oxalic acid. After ignition, the appearance of the oxides or a spectrographic analysis is used to determine whether a repetition of the treatment is necessary. Production data on the two latest neodymium series are shown in Table 10.

Silicon

The isotopes of silicon were collected in copper pockets in order to avoid the formation of the refractory silicon carbide. The acid-insoluble silicon is recovered by dissolving away part of the copper. After filtration, the silicon is converted to silicate by fusion with sodium carbonate. Hydrated silicic acid is obtained by dissolution in hydrochloric acid, evaporation to dryness, redissolving of the impurities in acid, and filtration. Ignition gives silicon dioxide, which may require retreatment with both alkali and acid. When quantities are small, perchloric acid may be substituted for the hydrochloric acid, with the advantage that quantitative yields of silicic acid are obtained by merely evaporating to strong fumes of the acid. Production data are given in Table 11.

Sulfur

Several methods of converting sulfides quantitatively to elemental sulfur have been studied, but because it has not been possible to give the necessary large effort to this difficult problem, the latest sulfur isotopes produced have been placed in the inventory as cadmium sulfide. The presence of so much cadmium outweighs the significance of all the impurities in the material.

Boron

The isotopes of boron are recovered from copper pockets and dissolved in the same manner as are the silicon isotopes. The boron is distilled as methyl borate after acidification with acetic acid and the addition of methanol. Hydrolysis with dilute hydrochloric acid gives boric acid, from which the liquids can all be distilled. Carry-over of impurities usually requires a repetition of both distillations. Information from the latest production of boron is given in Table 12.

Palladium

The isotopes of palladium are collected in carbon pockets, which are burned in an oxygen atmosphere and leave impure palladium metal. This is dissolved in nitric acid, diluted to slight acidity, and precipitated by an ethanol solution of dimethylglyoxime. The voluminous precipitate is then

[1]Summer employee.

TABLE 10. PRODUCTION DATA ON NEODYMIUM ISOTOPES

ISOTOPE	LOT NO.	WEIGHT OF Nd_2O_3 (g)	IMPURITIES FOUND (%)								
			Pr	Sm	Gd	Al	Ba	Ca	Fe	Mg	Si
142	FX738(a)	0.6153		<0.1					<0.04		
	FX738(b)	1.5297		<0.1					<0.04		
	GI783(a)	28.3278			<0.1			0.08			0.08
143	FX739(a)	0.3710		<0.1							
	FX739(b)	0.5914		<0.1							
	GI784(a)	12.5950			<0.1			0.08			0.2
144	FX740(a)	0.4400		<0.1							
	FX740(b)	1.0934					0.04				
	GI785(a)	18.8283								<0.01	
145	FX741(a)	0.5371		<0.1							
	GI786(a)	6.5096	None detected								
146	FX742(a)	0.5416	None detected								
	GI787(a)	11.9415	None detected								
148	FX743(a)	0.1232						<0.15		<0.02	0.08
	FX743(b)	0.0728					0.04	<0.15		<0.02	0.08
	GI788(a)	3.6245	<0.2	<0.2							
150	FX744(a)	0.1195				0.04	0.04			0.15	0.1
	GI789(a)	2.4160	<0.2	<0.2							

TABLE 11. PRODUCTION DATA ON SILICON ISOTOPES

ISO-TOPE	LOT NO.	WEIGHT OF SiO_2 (g)	IMPURITIES FOUND (%)									
			Ag	B	Co	Cu	Mg	Na	Pt	Sn	Ti	Zn
28	GC761(a)	20.5040			<0.05		0.001				0.01	0.05
29	GC762(a)	1.9415				<0.02	0.01	0.3		<0.05		
30	GC763(a)	0.5640	0.04	0.015		0.005	0.005		0.15			

TABLE 12. PRODUCTION DATA ON BORON ISOTOPES

ISOTOPE	LOT NO.	WEIGHT OF H_3BO_3 (g)	IMPURITIES FOUND (%)		
			Mg	Na	Si
10	GF768(a)	0.5726			0.08
11	GF769(a)	5.1097	0.01	0.04	0.02

ignited to form the metal. When quantities are too large for convenient handling of the dimethylglyoxime precipitate, ammonium chloropalladate is precipitated by addition of a considerable excess of ammonium chloride after oxidation of the palladium with chlorine. The salt is reduced to metal by heating. Production data are given in Table 13.

Platinum

Impure platinum is dissolved slowly from pocket ash by repeated treatments with aqua regia. All nitrate is then removed by numerous digestions with hydrochloric acid. The solution is diluted, and ammonium chloroplatinate is precipitated by addition of an excess of ammonium chloride in the presence of ethanol. The salt is reduced to the metal by heating. Production data are shown in Table 14.

SEPARATION OF RARE EARTHS

F. M. Scheitlin F. B. Thomas
F. A. Kappelmann A. C. Topp[2]

The liquid-liquid extraction process, which uses undiluted tributyl phosphate and nitric acid as the solvents, was continued so that quantities of purified rare earths could be produced for the separation of their isotopes. The supply of neodymium was augmented, and sufficient quantities of samarium and gadolinium were supplied for single, calutron production series. The first separation of gadolinium isotopes was made possible by these operations.

[2]Summer research participant, Georgia Institute of Technology, Atlanta, Georgia.

TABLE 13. PRODUCTION DATA ON PALLADIUM ISOTOPES

ISOTOPE	LOT NO.	WEIGHT OF Pd (g)	IMPURITIES FOUND (%)								
			Ag	Ca	Cu	Cr	Fe	Mg	Pt	Si	Ti
102	GG770(a)	0.3583	0.04	0.15			<0.04	0.02		0.08	<0.04
104	GG771(a)	1.8327			<0.02			<0.02		0.08	
105	GG772(a)	1.1475			<0.02	0.15	<0.04	0.02	Trace		
106	GG773(a)	2.9673	<0.04		<0.02			<0.02		<0.08	
108	GG774(a)	4.0269	<0.04	0.08	0.02			0.02		0.08	
110	GG775(a)	1.7484	<0.04	0.15	0.02			<0.02		<0.08	

TABLE 14. PRODUCTION DATA ON PLATINUM ISOTOPES

ISOTOPE	LOT NO.	WEIGHT OF Pt (g)	IMPURITIES FOUND (%)					
			Cu	Fe	Mg	Mn	Si	Ti
190	GH776(a)	0.3446	<0.01		0.01	<0.01	0.01	
192	GH777(a)	0.2789	0.02		0.005		0.02	
192	GH777(b)	0.9112			0.02			
194	GH778(a)	12.0060		0.05				
195	GH779(a)	3.2157	None detected					
196	GH780(a)	6.9895	None detected					
198	GH781(a)	0.4977	0.01	0.02	0.002		0.05	0.02
198	GH781(b)	1.0496	None detected					

The purities attained varied with the batches, but the samarium, gadolinium, and neodymium supplied for calutron operation contained from 97 to 99.5% of the desired element. Since samarium is the most abundant element in the raw material used, there is now on hand, as nitrate solution, the equivalent of more than 20 kg of Sm_2O_3 of 98 to 99.5% quality. A still larger quantity will be obtained from future operations directed toward separating europium from a mixture consisting mostly of samarium and gadolinium. The same operations, as well as the current purification of dysprosium, will also yield kilogram quantities of gadolinium. A considerable quantity of both these elements can be made available for uses other than isotope separation. A memorandum to division directors lists the charges which will be necessary. The capacity of the pulse columns now in use is such that additional enhancement of quality could be accomplished in a short time. Experience gained in the purification of the separated isotopes has shown that the additional efforts necessary to increase purity to 99.9% would have been justified.

An example of accomplishment with the present system was the separation of 99.5% samarium in a 90% yield from a mixture containing 40% samarium, 40% gadolinium, and 20% europium and heavy rare earths. The feed rate was about 400 g/hr.

The two pulse columns available have been joined, and this results in the expected increase in separation. A short, Rushton-type column is used to strip the rare earths from the TBP with 2 *N* nitric acid.

Satisfactory separations have been obtained between dysprosium and terbium when the pulse columns have been operated with nitric acid concentrations at 10.5 and 12.5 *N*. Still higher acidities have given good results in York-Scheibel columns. Two pulse columns have been joined vertically to give 22 mechanical stages. Gravity feed through rotameters has been replaced by the use of small pumps for all three liquids.

The separation between dysprosium and yttrium had been expected to be difficult, since yttrium falls between dysprosium and its nearest neighbor, holmium. The difficulty, however, has been greater than that anticipated. In nearly all operations with both types of columns, there has been persistent evidence of overlap of the heavier elements, with erbium sometimes being enriched in the aqueous solution rather than being completely removed from it. Interpretation of experimental data from batch extractions has offered an explanation of this phenomenon.

A large number of small, single-stage, batch extractions have been made from solutions of samarium and gadolinium nitrates of various concentrations. A few experiments have been performed with the small quantities of dysprosium and yttrium available. The following qualitative deductions can be made:

1. The distribution coefficient of each element in dilute solution is an exponential function of the nitric acid concentration.

2. The distribution coefficient of each element is an inverse function of the rare-earth concentration, only in solutions with the nitric acid concentration greater than a value specific for the element. The lighter elements have a greater independence of concentration.

3. The acidity at which the maximum separation factor between a pair of elements, the ratio of their distribution coefficients, is obtained varies with the elements. Thus, samarium and gadolinium separate best when extracted from 13 to 14 *N* nitric acid. The separation between gadolinium and terbium or gadolinium and dysprosium takes place best at a slightly lower acidity. The optimum acid concentration for removing yttrium from those elements which it most closely resembles may be as low as 8 or 9 *N*. Experiments are now under way to determine the proper flow ratios for the separation of dysprosium and yttrium at reduced acidity. It should be possible to eliminate the overlap of erbium and ytterbium, which can now be explained as the effect of rare-earth concentration on distribution coefficients at excessive acidities.

In spite of the difficulties, there is now on hand the equivalent of about 3 kg of dysprosium oxide, contaminated by only half as much yttrium and other rare earths.

ISOTOPE SHIPMENTS AND REQUESTS

One hundred fifty-two individual isotopes were shipped to research laboratories under allocations approved by the AEC. In comparison, 118 individual shipments were made for the six-month period ending May 20, 1953, and 115 individual shipments were made during the corresponding period from

May 20 to November 20, 1952. The following tabulation shows the distribution of shipments among the recipients:

Recipient	Number of Shipments
ORNL	20
Argonne	13
Brookhaven	18
Los Alamos	12
University of California	10
Other universities	71
All other government and industrial organizations	8
TOTAL	152

A tabulation of shipments made and the status of requests for allocations are given in Table 15.

PUBLICATIONS

Paper published August 20: Boyd Weaver, F. A. Kappelmann, and A. C. Topp, "Quantity Separation of Rare Earths by Liquid-Liquid Extraction. I. The First Kilogram of Gadolinium Oxide," *J. Am. Chem. Soc.* **75**, 3943 (1953).

Paper presented before the Division of Analytical Chemistry of the American Chemical Society at the 124th meeting, September 16, 1953: Boyd Weaver, "Fractional Separation of Rare Earths by Oxalate Precipitation from Homogeneous Solution."

TABLE 15. SUMMARY OF ISOTOPE SHIPMENTS AND REQUESTS, MAY 20 TO NOVEMBER 20, 1953

REQUEST NO.	REQUESTER	ISOTOPE	LOT NO.	QUANTITY (mg)
	Shipments Made			
B-3470	Bell Telephone Laboratories	Li^{6}	ED 537(a)	1
B-3471	Bell Telephone Laboratories	Cu^{65}	DU 498(a)	3
B-3472	Bell Telephone Laboratories	Ni^{61}	AS-AT 143-149(a)	1
B-3462	Florida State University	Mo^{95}	AW 165(ar)	183
B-3463	Florida State University	Mo^{100}	FP 701(a)	76
B-3540S	Oak Ridge National Laboratory	Ca^{44}	DI 444(a)	135
B-3541S	Oak Ridge National Laboratory	Ca^{44}	CS 373(a)	90
B-3525	University of California	Sr^{87}	EH 550(a)	1,001
B-3543S	Los Alamos Scientific Laboratory	Zr^{90}	EE 539(a)	2,006
B-3542S	Los Alamos Scientific Laboratory	Te^{120}	DX 505(a)	12
B-3551S	Oak Ridge National Laboratory	In^{113}	ER 594(a)	26
B-3568	University of Michigan	Ge^{76}	FZ 751(a)	200
B-3554	Ohio State University	Fe^{56}	CZ 404(f)	500
B-3544	University of Illinois	Cu^{63}	FJ 673(a)	400
B-3545	University of Illinois	Cu^{65}	DU 498(a)	400
B-3565	Massachusetts Institute of Technology	Li^{6}	FY 745(b)	114
B-3622S	Westinghouse Atomic Power Division	Li^{7}	FY 746(a)	5,500
B-3597S	Brookhaven National Laboratory	Hf^{176}	ET 603(a)	57
B-3596S	Brookhaven National Laboratory	Hf^{174}	ET 602(a)	14
B-3617S	Oak Ridge National Laboratory	Li^{6}	ED 537(a)	100
B-3524	University of California	Zr^{91}	CK 335(ar)	600
B-3534	University of California	Ni^{61}	FJ 671(a)	250
B-3582S	Los Alamos Scientific Laboratory	Sn^{116}	EC 530(a)	1,000
B-3583S	Los Alamos Scientific Laboratory	Sn^{117}	EC 531(a)	1,001
B-3546S	Argonne National Laboratory	Ni^{64}	AS-AT 146-152(a)	4
B-3613	Massachusetts Institute of Technology	Te^{124}	FE 643(a)	100
B-3612	Massachusetts Institute of Technology	Sn^{124}	EC 536(as)	100
B-3611	Massachusetts Institute of Technology	Mo^{96}	FP 698(a)	100
B-3590	University of Washington	B^{10}	FV 734(a)	500
B-3589	University of Washington	B^{11}	FV 735(a)	500
B-3588	Case Institute of Technology	Li^{6}	FY 745(b)	290
B-3372	Princeton University	Li^{6}	FY 745(a)	171
B-3618	Rice Institute	Cu^{65}	DU 498(a)	52
B-3634S	Brookhaven National Laboratory	Br^{79}	EJ 552(a)	50

TABLE 15. (continued)

REQUEST NO.	REQUESTER	ISOTOPE	LOT NO.	QUANTITY (mg)
	Shipments Made			
B-3635S	Brookhaven National Laboratory	Br^{81}	EJ 553(a)	51
B-3586	University of Michigan	Te^{122}	FE 641(a)	150
B-3587	University of Michigan	Te^{124}	CA 295(b)	225
B-3489	University of Michigan	Te^{130}	DX 512(a)	201
B-3661S	Oak Ridge National Laboratory	Sm^{152}	EM 573(a)	700
B-3668S	Oak Ridge National Laboratory	Fe^{58}	FN 668(d)	300
B-3669S	Oak Ridge National Laboratory	Fe^{58}	DO 480(b)	300
B-3147S	Iowa State College	Li^{6}	FY 745(b)	200
B-3620	University of Utah	Hg^{200}	DR 488(b)	44
B-3696S	Oak Ridge National Laboratory	Se^{74}	FG 654(a)	30
B-3630	Wesleyan University	Si^{30}	AI-AJ 105-108(a)	50
B-3721S	Oak Ridge National Laboratory	Ce^{136}	FR 706(a)	50
B-3722S	Oak Ridge National Laboratory	Ce^{138}	FR 707(a)	100
B-3710S	University of California Radiation Laboratory	Se^{74}	FG 654(a)	60
B-3734S	Los Alamos Scientific Laboratory	Ti^{50}	EN 580(a)	60
B-3699	National Bureau of Standards	Li^{6}	FY 745(b)	50
B-3753	Naval Medical Research Institute	In^{113}	DE 419(a)	1,000
B-3754	Naval Medical Research Institute	Sm^{152}	EM 573(a)	175
B-3727	Rice Institute	Li^{7}	FI 668(d)	50
B-3681S	Argonne National Laboratory	Fe^{57}	FC 636(a)	1,000
B-3680S	Argonne National Laboratory	Fe^{56}	CZ 404(f)	2,000
B-1878	Columbia University	Li^{6}	FY 745(b)	1,000
B-3719	Massachusetts Institute of Technology	Nd^{142}	EQ 587(a)	50
B-3718	Massachusetts Institute of Technology	Ce^{142}	FK 678(a)	100
B-3717	Massachusetts Institute of Technology	Cd^{112}	CE 318(a)	100
B-3716	Massachusetts Institute of Technology	Mo^{97}	R-T 57(a)	100
B-3715	Massachusetts Institute of Technology	Mo^{92}	FP 695(a)	50
B-3737S	Oak Ridge National Laboratory	Li^{7}	EZ 628(e)	1,000
B-3702S	Argonne National Laboratory	Li^{6}	FY 745(y)	1,000
B-3735	University of Wisconsin	Sm^{154}	FH 666(a)	200
B-3709	University of Chicago	K^{41}	EY 626(e)	40
B-3724S	Brookhaven National Laboratory	Sn^{117}	EC 531(a)	1,500
B-3725S	Brookhaven National Laboratory	Sn^{119}	EC 533(a)	1,500

TABLE 15. (continued)

REQUEST NO.	REQUESTER	ISOTOPE	LOT NO.	QUANTITY (mg)
	Shipments Made			
B-3744S	Argonne National Laboratory	Sb^{121}	AE 93(ar)	50
B-3738S	Argonne National Laboratory	V^{50}	FM 683(b)	35
B-3740S	Argonne National Laboratory	Zn^{70}	EK 560(a)	51
B-3743S	Argonne National Laboratory	Ti^{50}	EN 580(a)	50
B-3701S	Oak Ridge National Laboratory	Li^{6}	FY 745(y)	1,036
B-3755S	Los Alamos Scientific Laboratory	Li^{6}	GD 764(e)	11
B-3756S	Oak Ridge National Laboratory	Li^{6}	FY 745(y)	302
B-3829S	Brookhaven National Laboratory	Ge^{74}	FZ 750(a)	20
B-3798S	Oak Ridge National Laboratory	Li^{7}	FI 668(d)	14,220
B-3805	Massachusetts Institute of Technology	Zn^{64}	EK 556(a)	2,000
B-3779S	University of California Radiation Laboratory	Mg^{24}	BZ 288(a)	2,503
B-3778S	University of California Radiation Laboratory	Mg^{26}	DZ 521(a)	580
B-3782	Johns Hopkins University	Cl^{35}	FW 736(a)	700
B-3781	Johns Hopkins University	Rb^{87}	EW 620(a)	600
B-3783	Johns Hopkins University	Cl^{37}	FW 737(a)	600
B-3829S	Oak Ridge National Laboratory	Mo^{98}	AW 168(a)	200
B-3812	University of Michigan	Pd^{102}	GG 770(a)	30
B-3763	Washington University	K^{41}	FA 631(a)	300
B-3818S	Oak Ridge National Laboratory	Cd^{111}	AC 89(a)	101
B-3840S	Oak Ridge National Laboratory	Cd^{110}	CE 316(a)	101
B-3773	University of Wisconsin	Sn^{124}	EC 536(a)	30
B-3772	University of Wisconsin	Sn^{122}	EC 535(a)	250
B-3766	University of Wisconsin	Sn^{115}	FS 712(a)	63
B-3764	University of Wisconsin	Sn^{112}	EC 527(au)	88
B-3678S	Argonne National Laboratory	B^{11}	FV 735(a)	1,201
B-3679S	Argonne National Laboratory	Fe^{54}	FC 634(a)	325
B-3599	University of Michigan	Sm^{147}	FH 661(a)	150
B-3845S	Los Alamos Scientific Laboratory	Pd^{105}	GG 772(a)	600
B-3853S	University of California Radiation Laboratory	Li^{7}	EZ 606(e)	500
B-3852S	University of California Radiation Laboratory	Li^{6}	GD 764(e)	50
B-3654	Northwestern University	Li^{6}	FY 745(bb)	211
B-3896S	Brookhaven National Laboratory	Ga^{71}	EI 555(a)	300
B-3662S	Oak Ridge National Laboratory	Li^{6}	FY 745(bc)	3,000

TABLE 15. (continued)

REQUEST NO.	REQUESTER	ISOTOPE	LOT NO.	QUANTITY (mg)
	Shipments Made			
B-3790	Purdue University	Sb^{121}	AE 93(ar)	10
B-2500	University of Pennsylvania	Sn^{116}	EC 530(a)	20
B-3841S	Argonne National Laboratory	Ni^{61}	FJ 671(a)	5
B-3848S	Argonne National Laboratory	Pd^{102}	GG 770(a)	10
B-3849S	Argonne National Laboratory	Pd^{104}	GG 771(a)	10
B-3847	Princeton University	Sb^{121}	AE 93(ar)	50
B-3851	Massachusetts Institute of Technology	Fe^{54}	DO 477(a)	2,000
B-3703	University of Rochester	Li^{6}	FY 745(bb)	2,500
B-3869	Washington University	Ti^{47}	BF 216(ar)	100
B-3893S	Los Alamos Scientific Laboratory	Zn^{64}	EK 556(a)	501
B-3894S	Los Alamos Scientific Laboratory	Te^{122}	EX 506(a)	86
B-3762	Stanford University	Pb^{208}	BO 260(a)	200
B-3912S	Los Alamos Scientific Laboratory	Ga^{69}	EI 554(a)	51
B-3913S	Los Alamos Scientific Laboratory	Ga^{71}	EI 555(a)	50
B-3813	University of Michigan	Pt^{190}	GH 776(a)	25
B-3814	University of Michigan	Pt^{192}	GH 777(a)	25
B-3815	University of Michigan	Pt^{194}	GH 778(a)	25
B-3884	Carnegie Institution of Washington	K^{41}	EK 623(e)	20
B-3885S	Brookhaven National Laboratory	Hg^{196}	DR 485(c)	50
B-3886S	Brookhaven National Laboratory	Hg^{198}	DR 486(a)	50
B-3887S	Brookhaven National Laboratory	Hg^{199}	DR 487(a)	46
B-3888S	Brookhaven National Laboratory	Hg^{200}	DR 488(a)	52
B-3889S	Brookhaven National Laboratory	Hg^{201}	DR 489(b)	52
B-3890S	Brookhaven National Laboratory	Hg^{202}	DR 490(c)	51
B-3891S	Brookhaven National Laboratory	Hg^{204}	DR 491(b)	51
B-3883	Florida State University	Cr^{54}	CD 312(a)	350
B-3860	Carnegie Institute of Technology	La^{138}	EF 544(a)	501
B-3861	Carnegie Institute of Technology	Nd^{144}	FX 740(b)	500
B-3864	Carnegie Institute of Technology	La^{139}	EF 545(a)	8,001
B-3911S	Carbide and Carbon Chemicals Co.	Li^{6}	FY 745(y)	1,500
B-3930	Yale University	Ca^{40}	EV 613(a)	201
B-3931	Yale University	Ca^{42}	EV 614(a)	100
B-3932	Yale University	Ca^{43}	FO 691(a)	125
B-3933	Yale University	C^{44}	FF 651(a)	150

TABLE 15. (continued)

REQUEST NO.	REQUESTER	ISOTOPE	LOT NO.	QUANTITY (mg)
	Shipments Made			
B-3831	Louisiana State University	Ba^{130}	FU 727(a)	10
B-3902	University of Texas	B^{11}	FV 635(a)	200
B-3062	University of Illinois	Si^{29}	EB 525(a)	86
B-3953	Columbia University	Zn^{64}	EK 556(a)	800
B-3966S	Brookhaven National Laboratory	Sn^{120}	EC 534(a)	2,001
B-3965S	Brookhaven National Laboratory	Sn^{116}	EC 530(a)	2,002
B-3964S	Brookhaven National Laboratory	Sn^{118}	FS 715(a)	2,002
B-3952	Carnegie Institute of Technology	W^{180}	EL 561(a)	101
B-3926	University of Rochester	Cd^{106}	CE 314(a)	20
B-3267	University of California at Los Angeles	Si^{29}	GC 762(a)	200
B-3961	Harvard University	Tl^{205}	CJ-3 333(a)	1,700
B-3967	University of Wisconsin	Mo^{92}	R-T 53(a)	45
B-3968	University of Wisconsin	Mo^{94}	FP 696(a)	75
B-3969	University of Wisconsin	Mo^{96}	FP 698(a)	100
B-3975S	Oak Ridge National Laboratory	Zn^{68}	EK 559(a)	750
B-3972S	Los Alamos Scientific Laboratory	Sn^{112}	FS 710(a)	125
B-3955	University of California	Hg^{202}	DR 490(a)	20

Requests on Hand or Held by AEC as of November 20, 1953

REQUEST NO.	REQUESTER	ISOTOPE	QUANTITY DESIRED (mg)
Awaiting purchase order			
B-3398	Duke University	K^{41}	840
B-3979	Duke University	Li^{6}	50
B-3993	Massachusetts Institute of Technology	Re^{185}	100
B-3994	Massachusetts Institute of Technology	Re^{187}	100
B-3995	Massachusetts Institute of Technology	Hg^{198}	100
B-3996	Massachusetts Institute of Technology	Hg^{199}	30
B-3997	Massachusetts Institute of Technology	Hg^{200}	100
B-3998	Massachusetts Institute of Technology	Hg^{201}	25
B-3999	Massachusetts Institute of Technology	Hg^{202}	100
B-4000	Massachusetts Institute of Technology	Hg^{204}	100
B-4001	Massachusetts Institute of Technology	W^{182}	100
B-4002	Massachusetts Institute of Technology	W^{183}	100
B-4003	Massachusetts Institute of Technology	W^{184}	100

TABLE 15. (continued)

Requests on Hand or Held by AEC as of November 20, 1953

REQUEST NO.	REQUESTER	ISOTOPE	QUANTITY DESIRED (mg)
B-4004	Massachusetts Institute of Technology	W^{186}	100
B-4006	University of Wisconsin	Se^{76}	75
B-4007	University of Wisconsin	Se^{77}	25
B-4008	University of Wisconsin	Se^{82}	50
B-4009	University of Wisconsin	Mo^{98}	100
B-4010	University of Wisconsin	Mo^{100}	25
B-4011	Argonne National Laboratory	Pb^{206}	10
Awaiting sample preparation			
B-4014S	Oak Ridge National Laboratory	Li^{6}	12,000
B-3650	Westinghouse Research Laboratory	Li^{6}	500
Requests for isotopes which have not been separated			
S-343	Oak Ridge National Laboratory	$Dy^{158,160}$	10 each
S-361	Argonne National Laboratory	Os^{192}	10
Requests for isotopes which have been separated but chemical refinement incomplete			
B-3862	Oak Ridge National Laboratory	Ru^{96}	200
B-3903	Argonne National Laboratory	Ru^{96}	10
B-3904	Argonne National Laboratory	Ru^{98}	10
S-148	Argonne National Laboratory	$Ir^{191,193}$	30 each
B-2333	Princeton University	Gd^{152}	50
B-2556S	University of California	Gd^{154}	10
B-2577S	University of California	Gd^{152}	10
B-3069	University of Michigan	Gd^{158}	100
B-3290	Yale University	Gd^{152}	100
B-3291	Yale University	Gd^{154}	100
B-3862	Carnegie Institute of Technology	Gd^{152}	1,000
Old requests for isotopes now available (require confirmation from requester)			
B-297	Argonne National Laboratory	$Pd^{102,104}$ $Pd^{105,106}$ $Pd^{108,110}$	200 each
B-269	Princeton University	B^{11}	500
Requests for isotopes recently added to the inventory, awaiting AEC approval			
B-3062	University of Illinois	Si^{29}	100

TABLE 15. (continued)

Requests on Hand or Held by AEC as of November 20, 1953

REQUEST NO.	REQUESTER	ISOTOPE	QUANTITY DESIRED (mg)
B-3267	University of California	Si^{29}	200
B-3269	University of California	S^{33}	200
B-3917	Stanford University	S^{33}	50
Requests awaiting replenishment of inventory			
B-2581	Argonne National Laboratory	Mo^{94}	600
B-3249	Brookhaven National Laboratory	Ni^{64}	50
B-3954	Columbia University	Ni^{64}	100
B-3398	Duke University	K^{41}	3,000
B-3872	Columbia University	K^{41}	1,000
B-3897	Yale University	Ca^{48}	100
B-3018	University of Rochester	Zr^{96}	200
B-3222	Massachusetts Institute of Technology	Zr^{96}	100
B-3417	Massachusetts Institute of Technology	Zr^{96}	250
B-3780	Johns Hopkins University	Rb^{85}	700
B-3467	Yale University	Hf^{177}	50
B-3468	Yale University	Hf^{179}	50
B-3988	Massachusetts Institute of Technology	Hf^{176}	100
B-3989	Massachusetts Institute of Technology	Hf^{177}	100
B-3990	Massachusetts Institute of Technology	Hf^{178}	100
B-3991	Massachusetts Institute of Technology	Hf^{179}	100
B-3992	Massachusetts Institute of Technology	Hf^{180}	100
B-3934	University of Wisconsin	Sm^{144}	250
B-3936	University of Wisconsin	Sm^{148}	250
B-3937	University of Wisconsin	Sm^{149}	250
B-3938	University of Wisconsin	Sm^{150}	250
B-3939	University of Wisconsin	Sm^{152}	250
B-3940	University of Wisconsin	Sm^{154}	250
B-3935	University of Wisconsin	Sm^{147}	250
B-3960	Harvard University	Tl^{203}	1,700
B-4022	Ohio State University	Fe^{54}	1,000
B-4025	University of Kentucky	Li^{6}	500