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QUARTERLY PROGRESS REPORT

STABLE ISOTOPE RESEARCH AND PRODUCTION DIVISION

OCTOBER 1, 1951 to DECEMBER 31, 1951

AEC RESEARCH AND DEVELOPMENT REPORT

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

[redacted]

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This document contains 38 pages.
This is copy 34 of 74 ,Series A.

Subject Category: Isotope
Separation

QUARTERLY PROGRESS REPORT
Stable Isotope Research and Production Division

October 1, 1951 to December 31, 1951

C. P. Keim, Director

J. R. Walton, Editor

January 2, 1952

[redacted]

OAK RIDGE NATIONAL LABORATORY
Operated by
CARBIDE AND CARBON CHEMICALS COMPANY
A Division of Union Carbide and Carbon Corporation
Oak Ridge, Tennessee

Contract No. W-7405-eng-26

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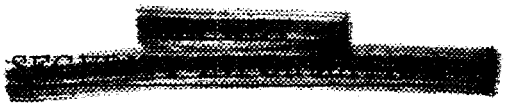

Stable Isotope Research and Production Division quarterly progress reports previously issued in this series are as follows:

Y-497 July 1, 1949 to September 30, 1949.

Y-550 October 1, 1949 to December 31, 1949.

Y-586 January 1, 1950 to March 31, 1950.

Y-636 April 1, 1950 to June 30, 1950.

Y-675 July 1, 1950 to September 30, 1950.

Y-723 October 1, 1950 to December 31, 1950.

Y-765 January 1, 1951 to March 31, 1951.

Y-797 April 1, 1951 to June 30, 1951.

Y-821 July 1, 1951 to September 30, 1951.

[redacted]

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STABLE ISOTOPE SEPARATION RESEARCH AND CALUTRON OPERATIONS

C. E. Normand, Department Head

INTRODUCTION

During the second quarter of fiscal year 1952, the Isotope Production and Process Development Department placed primary emphasis on the collection of lithium, iron and calcium isotopes.

COLLECTIONS

Three elements were processed and twelve isotopically enriched samples were collected.

Lithium, Series F1-XAX

(L. O. Love, W. A. Bell, C. V. Ketron, G. J. Fisher, R. L. Caldwell, K. A. Spainhour, and operating personnel)

Lithium isotopes (Li6 and Li7) have been collected in the Alpha channels. There have been no changes in the equipment as reported in "Progress Report, Stable Isotope Research and Production Division," Y-821.

Innage was reduced during a two-week period while the pumping system was being modified. Small water leaks also developed that were extremely difficult to locate.

The lava anode insulator disintegrated during operation with the use of the mixed type of charge material (lithium bromide and lithium metal). Lithium fluoride insulators, prepared in the ceramics laboratory of Y-12 plant, were substituted for the lava and these proved satisfactory under operating conditions.

Iron, Series FN-XBX (L. O. Love, W. A. Bell, C. V. Ketron, G. J. Fisher, R. L. Caldwell, K. A. Spainhour, and operating personnel)

This collection of iron isotopes (Fe54, Fe56, Fe57, and Fe58) was started in June in the Beta channels and was completed in November. Main efforts were directed toward collecting a sample containing a high percentage of Fe58 in relation to Fe54. No changes have been made in equipment or operating techniques since last reported.[1] Receiver pocket erosion was the major collection problem throughout this series. Persistent electrical shorts between the pockets was another source of trouble.

Sixty of the 110 runs made during this series were terminated for filament failure. The average innage time for these runs was 34.6 hours. Innage time for the remaining 50 runs averaged 21.9 hours. This figure includes 12 runs in which no innage was obtained. The total innage for the entire series was 3173 hours. The average total monitored beam received during this time was 100.7 ma. Sixteen separate samples were submitted to the Isotope Chemistry Department and the total collection estimated from monitored currents was as follows:

Fe54	30.86 grams
56	621.82 grams
57	12.77 grams
58	2.00 grams
Total	667.45 grams

Calcium, Series FO-XBX

The collection of calcium isotopes (Ca40, Ca42, Ca43, Ca44, Ca46, and Ca48) was started in November in the Beta channels and is being continued.

A medium temperature Beta source was used, embodying a deep floating anode type ionization chamber with a 3/16-inch zero degree faced, stainless steel ion exit slit. The filament was made of 170-mil tantalum. Calcium metal was used as the charge material. The operating temperature range was 650°-800°C as indicated by the thermohm, with best operation at approximately 700°C. This temperature is near the upper limit of the medium temperature source; therefore stainless steel heat shielding was added to both the charge oven and manifold castings to reduce heat losses.

1. "Quarterly Progress Report, Stable Isotope Research and Production Division, July 1, 1951 to September 30, 1951." Y-821.

All the beams were collected in water-cooled copper pockets placed behind 1/8-inch defining slots milled in the graphite defining faceplate. The slot at the mass 40 position was enlarged to 1/4 inch for the collection of the high natural abundance (96.96 per cent) Ca40 beam.

The formation of nitrides along with carbides within the ionization chamber as the run progressed created unstable arc conditions and lowered the output. By using helium instead of nitrogen as a support gas, deposits within the ionization chamber were reduced and both output and run stability were improved. The greatest difficulty at the present time arises from electrical shorting of the reflecting anode by deposits of calcium on nearby cold surfaces.

Electrical shorts at the Ca40 pocket were prevalent and were caused by the peeling off of material from the back of the defining faceplate in such a way as to touch the pocket. This peeling off was retarded by a number of holes drilled half way through the back of the faceplate around the Ca40 defining slot.

At this time 49 runs have been completed with an average innage time of 23 hours. Twenty-eight of these runs were terminated because of open filaments.

The average total ion current received during innage was 88 ma. The estimated total collection is as follows:

Ca40	143.39 grams
42	0.61 grams
43	0.25 grams
44	2.41 grams
46	0.01 grams
48	0.17 grams
Total	146.84 grams

PUBLICATIONS ISSUED AND IN PROGRESS

Publications

During the quarter, 11 reports appeared in the unclassified literature in which enriched stable isotopes had been used; 6 were university reports, 3 AEC project reports, and 2 jointly issued by university and AEC. Since stable isotopes were first distributed, 138 university reports, 66 project reports, and 36 reports from this division have been published in technical journals, all having used stable isotopes in their research.

Reports Issued

1. Stable Isotope Research and Production Division, "Progress Report of Lithium Activities," July 1, 1951 to October 31, 1951, ORNL-1166.
2. "A Survey of Calutron Charge Materials for Stable Isotope Separations," by C. P. Keim, Y-822.

Reports in Progress

1. Stable Isotope Separations in Y-12.

STABLE ISOTOPE CHEMISTRY, RESEARCH AND PRODUCTION

Boyd Weaver, Department Head

INTRODUCTION

The chief production functions of the Isotope Chemistry Research and Production Department are the supply of suitable charge materials for isotope separation by the calutrons, chemical refinement of enriched isotopes and the preparation of materials for their mass analysis. Chemical services and unusual compounds are also supplied to other departments of the Stable Isotope Research and Production Division.

The department carries on research activities in various fields related to isotope production and use, with recent interest centered around the elements lithium, zirconium, hafnium, and the rare earths.

All refined isotopes are stored in the chemistry depositories, and the preparation of all samples for shipment is performed by personnel of this department. Unused portions and many processed residues of isotopes are returned by research institutions for restoration in the laboratory's stock of isotopes. Some of these require reprocessing to make them suitable for reuse; others are too small quantities for further handling.

QUARTERLY SAMPLE SUMMARY

Isotope samples on which refinement was completed	21
Samples in process of chemical refinement, Dec. 31	14
Samples awaiting chemical refinement, Dec. 31	9
Samples submitted to Mass Analysis Laboratory	29
Mass Analyses reported	19
Samples awaiting mass analysis, Dec. 31	32
Stable Isotope shipments made	77

PRODUCTION OF CHARGE MATERIALS

Preparation and Purification (C. W. Sheridan and C. F. Harrison)

Charge materials of lithium, iron and calcium were prepared for calutron operations. Various chemical services were also supplied in those operations.

Because of difficulties in purchasing pure cerous chloride, crude cerium oxide was purified by crystallization of ammonium hexanitrato cerium and extraction of cerium (valence of 4) nitrate from nitric acid with diethyl ether. Both methods gave excellent separation from all the other rare earths. Repeated extractions were required because of reduction of cerium (4) in solution.

ISOTOPE REFINEMENT

Special Projects (W. C. Davis, R. L. Bailey, F. B. Thomas, K. A. Allen, and F. M. Scheitlin)

The major efforts of the department were devoted to special production and research projects which will be reported in a special report.

Iron (F. B. Thomas and F. M. Scheitlin)

Refinement of all samples of Fe58 in a prolonged separation project was completed.

Strontium (K. A. Allen)

Refinement of one of a series of strontium isotopes was completed.

ACTIVITIES ASSOCIATED WITH MASS ANALYSIS

Compound Preparation (F. M. Scheitlin)

Isotopes of six elements were prepared in suitable form for mass analysis. The elements and the forms in which they were submitted to the Mass Analysis Laboratory were as follows:

Element	Form
Lithium	LiI
Boron	H_3BO_3
Iron	$FeCl_3$
Sulfur	CdS
Cerium	CeO_2
Calcium	$CaCO_3$

SEPARATION OF RARE EARTHS

Extraction for Nitric Acid by Tributyl Phosphate (F. A. Kappelmann)

Work was continued on the separation of gadolinium from the other rare earths by the process described in Y-821, pp. 16 and 17. All of the neodymium and nearly all of the samarium have been separated from the other earths in a gadolinium concentrate with only small gadolinium loss. The next step will be to split gadolinium from yttrium and the heavier earths. At present a pseudo-countercurrent batch system with 9 stages on each side of the double reflux is being used. The only difficulty encountered with the process comes from the fact that separation factors are dependent on rare earths concentrations. Therefore more stages are necessary than were anticipated, but not a prohibitive number.

SPECIAL COMPOUND PREPARATION

Vanadium (F. M. Scheitlin)

For investigation of response to nuclear induction it was desirable to have approximately 300 milligrams of V50 in the form of the compound vanadium oxytrichloride ($VOCl_3$). This volatile liquid is produced by chlorination of vanadium trioxide (V_2O_3). Since about one-third of the vanadium is converted to vanadium pentoxide (V_2O_5) in a side reaction, the process must be repeated to obtain a high yield. After experiments with natural vanadium, the enriched isotope was converted to the desired compound with a yield of 92 per cent and no detectable loss.

ISOTOPE SHIPMENTS AND REQUESTS

Preparation for Shipment

Seventy-seven individual samples were prepared for shipment to laboratories within and outside of the Atomic Energy Commission. Several samples returned by users were also processed for further usage. Details of the shipments during the quarter and the present status of requests for isotopes are given in Table I.

Stable Isotope Sales (Fees)

On the basis of the nominal charge of $50.00 made to most users of stable isotopes for individual shipments, the following summary has been compiled:

	July 1 to Sept. 30, 1951	October 1 to Dec. 31, 1951
Transfers within AEC	$ 3,600.00	$ 2,250.00
Cash Sales (fees)	1,250.00	1,600.00
Total Sales (fees) and Transfers to Date	$66,150.00	$70,000.00

TABLE I

Shipments of Stable Isotopes during October, November December, 1951

Request Serial No.	Requestor	Isotope	Series No.	Sample No.	Quantity Shipped(mg)	Date Shipped
B-1779	Kansas State College	Sm149	FH	663(a)	10.1	10-18-51
B-2076S	Los Alamos Scientific Laboratory	Li6	FI	667(m)	1.0	10-17-51
B-2134S	University of California	S33	ES	597(a)	10.4	10-18-51
B-2173	University of Rochester	K40	EX	622(u)	10.5	10-31-51
B-2174	University of Rochester	Sn124	EC	536(a)	150.0	10-31-51
B-2175	University of Rochester	Zr96	EE	543(a)	50.0	10-31-51
B-2196	Rutgers University	Sn122	EC	525(a)	251.8	10-3-51
B-2201	University of Wisconsin	Mg24	DZ	519(a)	500.0	10-18-51
B-2202	University of Illinois	Sb121	BT	274(a)	11.3	10-18-51
B-2203	University of Illinois	Li7	CU	379(a)	2,001.0	10-18-51
B-2213S	University of California	Fe54	BA	187(ar)	75.8	10-18-51
B-2222	Yale University	Ti46	EN	576(a)	301.3	10-30-51
B-2223	Yale University	Ti47	BF	216(ar)	86.3	10-30-51
B-2224	Yale University	Ti48	EN	578(a)	502.0	10-30-51
B-2225	Yale University	Ti49	EN	579(a)	100.4	10-30-51
B-2226	Yale University	Ti50	BF	219(ar)	75.8	10-30-51
B-2228S	Oak Ridge National Laboratory	Ca42	DI	442(b)	200.0	10-31-51
B-2229S	Brookhaven National Laboratory	Sn124	EC	536(a)	2,220.5	10-11-51
B-2255	Naval Research Laboratory	Li6	FI	667(a)	20.4	10-30-51
B-1153	University of Wisconsin	Ce136	FK	675(a)	10.3	11-21-51
B-1896	University of Michigan	Sn124	EC	536(as) metal	800.0	11-20-51
B-1932S	Ames Laboratory(reshipment)	S33	ES	597(a)	208.8	11-5-51
B-2017	Princeton University	Sn124	EC	536(as) metal	508.2	11-20-51
B-2176S	Argonne National Laboratory	Li6	FI	667(a)	50.7	11-7-51
B-2195S	Argonne National Laboratory	Li6	FI	667(a)	100.0	11-7-51
B-2196	Rutgers University	Sn122	EC	535(a) metal	251.8	11-26-51
B-2258S	Argonne National Laboratory	Hg200	DR	488(a)	103.7	11-21-51
B-2259S	Argonne National Laboratory	Li6	FI	667(a)	100.1	11-21-51
B-2267	Washington University	Li7	BQ	265(ar)	504.9	11-12-51
B-2268	Northwestern University	Li6	FI	667(a)	100.1	11-12-51
B-2300S	Oak Ridge National Laboratory	Ca40	DI	441(d)	100.2	11-23-51
B-2301S	Oak Ridge National Laboratory	Cr50	EU	609(a)	76.2	11-23-51
B-2302S	Oak Ridge National Laboratory	Cr54	EU	612(a)	20.3	11-23-51
B-2303S	Oak Ridge National Laboratory	Fe54	DO	477(a)	2,000.1	11-23-51
B-2304S	Oak Ridge National Laboratory	Fe56	CZ	404(f)	2,000.0	11-23-51
B-2305S	Oak Ridge National Laboratory	Mg26	BZ	290(a)	100.6	11-23-51
B-2306S	Oak Ridge National Laboratory	Ni58	DF	422(a)	700.9	11-23-51
B-2307S	Oak Ridge National Laboratory	Ni60	DF	423(a)	40.5	11-23-51
B-2308S	Oak Ridge National Laboratory	Ni62	DF	425(a)	22.0	11-23-51
B-2310S	Oak Ridge National Laboratory	Ti46	BF	215(ar)	99.8	11-23-51

Request Serial No.	Requestor	Isotope	Series No.	Sample No.	Quantity Shipped(mg)	Date Shipped
B-2311S	Oak Ridge National Laboratory	Ti48	EN	578(a)	1,000.6	11-23-51
B-2312S	Oak Ridge National Laboratory	Ti50	EN	580(a)	50.6	11-23-51
B-2321S	Oak Ridge National Laboratory	Fe58	FN	688(e)	196.3	11-20-51
B-2322S	Oak Ridge National Laboratory	Fe58	FN	688(h)	165.9	11-20-51
B-2287S	Los Alamos Scientific Laboratory	Cd116	CE	321(ar)	451.7	11-27-51
B-2288S	Los Alamos Scientific Laboratory	Sn124 metal	EC	536(as)	502.7	11-27-51
B-2338S	Oak Ridge National Laboratory	Ce140	DW	503(a)	8,532.2	12-4-51
B-2339S	Oak Ridge National Laboratory	Nd142	EQ	587(a)	1,802.6	12-4-51
B-2340S	Oak Ridge National Laboratory	Nd144	EQ	589(a)	1,863.4	12-4-51
B-2336S	University of California	Nd145	EQ	590(a)	40.3	12-4-51
B-1963S	Vanderbilt University	Ce136	FK	675(a)	5.4	12-7-51
B-1410S	Brookhaven National Laboratory	Ca48	FF	653(a)	25.5	12-11-51
B-2309S	Oak Ridge National Laboratory	Sn124 metal	EC	536(as)	202.2	12-11-51
B-2250	Bartol Research Foundation	Sn124 metal	EC	536(as)	150.2	12-11-51
B-2354	Indiana University	Ge74	BD-BE	208-213(a)	500.1	12-13-51
B-2335S	Brookhaven National Laboratory	K41	FA	631(a)	100.2	12-13-51
B-2325S	Argonne National Laboratory	Mg204	DR	491(b)	27.7	12-14-51
B-2332S	Argonne National Laboratory	Mg26	DZ	521(a)	30.3	12-14-51
B-2326S	Argonne National Laboratory	W186	EL	565(a)	50.0	12-14-51
B-2330S	Argonne National Laboratory	Si30	AI-AJ	105-108(a)	10.5	12-14-51
B-2329S	Argonne National Laboratory	Ti50	EN	580(a)	30.0	12-14-51
B-2328S	Argonne National Laboratory	Cd116	CE	321(ar)	30.2	12-14-51
B-2327	Argonne National Laboratory	Hf180	ET	607(a)	30.7	12-14-51
B-2331S	Argonne National Laboratory	Ca48	FF	653(a)	10.2	12-14-51
B-2350	Naval Research Laboratory	Cr53	EU	611(a)	401.0	12-17-51
B-2349	Naval Research Laboratory	Fe57	EN	471(a)	453.8	12-17-51
B-2027	Yale University	Sn124 metal	EC	536(as)	137.2	12-17-51
B-2314	Harvard University Medical School	Cr50	CD	309(ar)	100.1	12-17-51
B-2371S	Brookhaven National Laboratory	Cr54	EU	612(a)	101.0	12-18-51
B-2239	Air Force Cambridge Research Center	Tl205	BU-3	277(a)	500.2	12-20-51
B-2240	Air Force Cambridge Research Center	Cd114	CE	320(ar)	501.7	12-20-51
B-2237	Air Force Cambridge Research Center	Hg198	DR	486(a)	260.5	12-20-51
B-2238	Air Force Cambridge Research Center	Hg202	DR	490(f)	415.7	12-20-51

Unfilled Requests on Hand December 31, 1951

Requests for isotopes which have not been separated:

Request Number	Date	Requestor	Isotope	Quantity Desired (mg)
S-148	10-21-47	Argonne National Laboratory	Ir191	20-30
S-148	10-21-47	Argonne National Laboratory	Ir193	300
S-297	10-2-49	Argonne National Laboratory	Pd102, 104, 105, 106, 108, 110	200 ea.

Request Number	Date	Requestor	Isotope	Quantity Desired (mg)
S-342	3-3-49	Oak Ridge National Laboratory	Cd158, 160	10 ea.
S-343	3-3-49	Oak Ridge National Laboratory	Dy158, 160	10 ea.

Requests requiring replenishment of inventory (quality or quantity insufficient):

S-325	3-2-49	Oak Ridge National Laboratory	C137	3,000
S-328	3-2-49	Oak Ridge National Laboratory	B11	10,000

Requests awaiting purchase orders:

B-849	8-26-49	Ohio State University	Ca48	10
B-1603	1-2-51	Ohio State University	Sm144	100
B-1604	1-2-51	Ohio State University	Sm147	50
B-2035	6-26-51	Massachusetts Institute of Technology	K40	15
B-2290	11-15-51	Ohio State University	Ca48	100
B-2373	12-17-51	Princeton University	Se82	400

Requests with Purchase Order awaiting replenishment of inventory:

B-2058S	7-10-51	Los Alamos Scientific Laboratory	Li6	50
B-2059S	7-10-51	Los Alamos Scientific Laboratory	Li7	10

Requests held by AEC Isotopes Division:

S-361	Argonne National Laboratory	Os192	10 mg
S-483	Brookhaven National Laboratory	Ba132	100
S-581	Brookhaven National Laboratory	K40	10
B-397	University of Wisconsin	Ca46	10-1,000
B-736	University of Chicago	Ca46	10
B-875	University of Illinois	Ba132	10
B-1062	Kansas State College	K40	1,000
B-1161	Princeton University	K40	150
B-1191	University of Wisconsin	V50	100
B-1490S	Argonne National Laboratory	Ru99	5
B-1491S	Argonne National Laboratory	Ru101	5
B-1601S	Brookhaven National Laboratory	V50	40
B-1696	Naval Medical Research Laboratory	Zn66	3,000
B-1709S	Argonne National Laboratory	Ni64	300
B-1805S	Argonne National Laboratory	Li7	200
B-1819	University of Illinois	Ca136	10
B-1915S	Brookhaven National Laboratory	Ca46	25
B-1963	Vanderbilt University	Ce136	20

Request Number	Requestor	Isotope	Quantity Desired (mg)
B-2007S	Brookhaven National Laboratory	Ba132	10
B-2309S	Oak Ridge National Laboratory	Sn124	400
B-2323	University of Pittsburgh	Cu63	3 g
B-2324	University of Pittsburgh	Cu65	3 g
B-2333	Princeton University	Gd152	50
B-2352	University of California	Hg204	100
B-2376	Washington University School of Medicine	Fe58	10
B-2392	Ohio State University	Ba130	100
B-2396	Argonne National Laboratory	Ge76	50
B-2401	Argonne National Laboratory	Nd150	20

ISOTOPE ANALYSIS METHODS LABORATORY

J. R. McNally, Jr., Department Head

NUCLEAR RESONANCE

Vanadium, $_{23}V^{50}$ (H. E. Walchli, W. E. Leyshon, and F. M. Scheitlin *)

An enhanced sample containing an estimated 25 mg of the odd-odd isotope $_{23}V^{50}$ was converted to vanadium oxytrichloride ($VOCL_3$), distilled into a spherical glass vial, sealed off, and then immersed in normal $VOCL_3$ as shown in Figure 1. The total chemical yield (three conversions) was 92 per cent efficient giving approximately 0.49 cc of $VOCL_3$. The nuclear resonance of $_{23}V^{50}$ was detected in this sample with a signal to noise ratio of greater than 50 to 1 (see Figure 2), and subsequently found in normal material at about three times noise. Frequency measurements with respect to the deuterium resonance gave

$$\nu(V^{50})/\nu(D^2) = 0.649527 \pm 0.00007$$

and

$$\left(\frac{\mu'}{I}\right)_{V_{50}} = +0.55690 \pm 0.00006,$$

the latter being uncorrected for nuclear diamagnetism. The Mayer shell model predicts both odd nucleons to be in $^2F_{7/2}$ states. If we assume the spins to be additive, the computed nuclear magnetic moment (corrected) becomes

$$\mu(V_{50}) = +3.9050 \text{ n. m.},$$

which may be compared to the Jj coupling value of 3.85 nuclear magnetons.

Technetium, $_{43}Tc^{99}$ (H. E. Walchli and R. Livingston**)

A sample containing 156 mg of Tc99, as NH_4TcO_4 in aqueous solution, was investigated for the nuclear resonance of technetium. The following results were obtained:

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* Stable Isotope Chemistry Research and Production Department.
** Chemistry Division, X-10 Area.

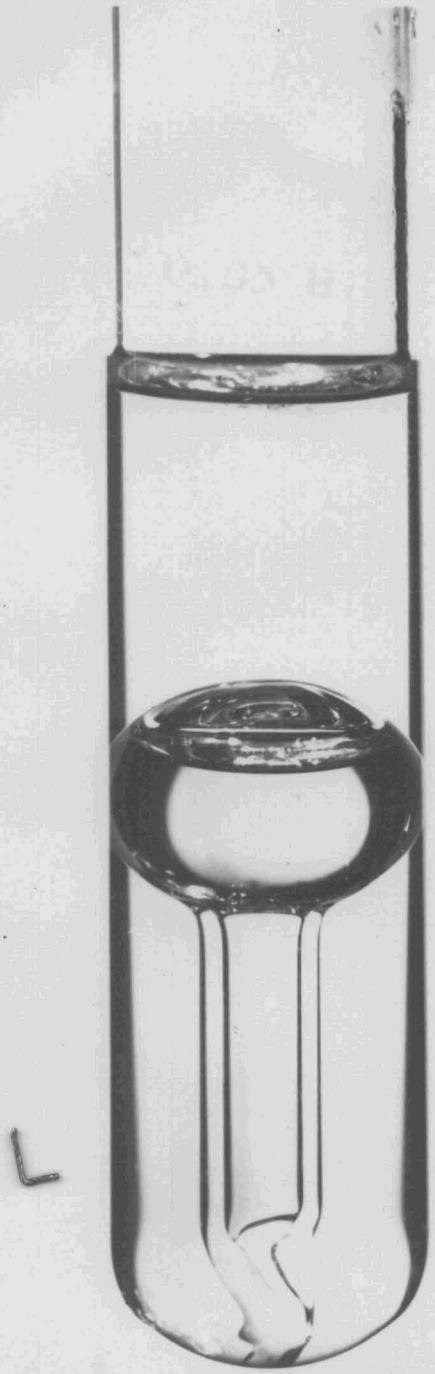

FIGURE 1 ENHANCED V^{50} SAMPLE AS $VOCl_3$ IN NORMAL $VOCl_3$ FOR NUCLEAR RESONANCE STUDY

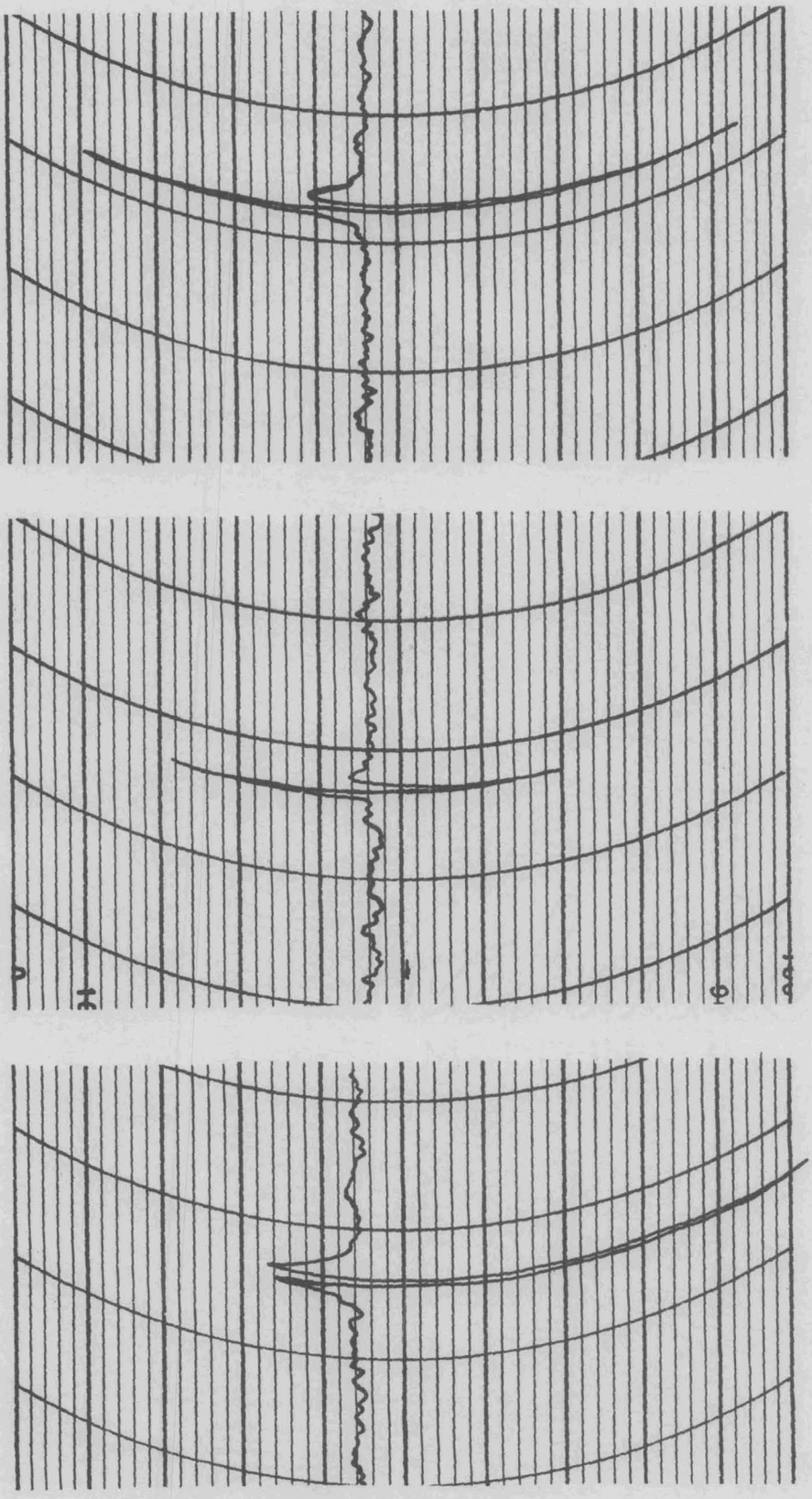

FIGURE 2 RECORDER TRACES

NUCLEAR RESONANCE

$$\nu(Tc^{99})/\nu(D^2) = 1.46628 \pm 0.0001$$

$$\mu(Tc^{99}) = +5.6805 \pm 0.0004 \text{ n.m.},$$

the latter value being corrected for nuclear diamagnetism.

Miscellaneous (H. E. Walchli)

New values of the resonance frequencies of Rb85 and Cl35 were also obtained and give, relative to the proton frequency (see Figure 2)

$$\nu(Rb^{85})/\nu(H^1) = 0.096554 \pm 0.00001$$

$$\nu(Cl^{35})/\nu(H^1) = 0.097985 \pm 0.00001.$$

Search for resonances in Si29, Se77, and U235 (as solid UF_6 only) has been unsuccessful so far. However, the sign and magnitude of the nuclear magnetic moment of P31 are currently being determined.

MICROWAVE SPECTROSCOPY

Microwave Spectrum of Vinyl Iodide (H. W. Morgan and J. H. Goldstein*)

Progress on the microwave spectrum of vinyl iodide has been slow because of equipment breakdown. A coaxial line tripler system (270 mc to 810 mc) employing a lighthouse tube has been put in operation in order to obtain useful reference frequencies in the 32,000 mc region.

Quadrupole Absorption Studies (H. W. Morgan)

The possibility of utilizing direct quadrupole transitions for detection of isotopes is under study. A regenerative detector circuit was constructed and Cl35 signals observed visually on a CRO with signal six times noise and only wide band amplification. Preliminary studies with both frequency and amplitude modulation and narrow band amplification gave enormous gains in signal strength. At present, it is estimated that about 25 mg of Cl35 can be detected by this method.

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* Consultant, Emory University, Atlanta, Georgia.

SPECTRO-ISOTOPIC ASSAY

Lithium (D. D. Smith, G. L. Stukenbroeker, and C. E. Pepper)

Approximately 100 standards and samples of lithium (as Li_2SO_4) have been assayed using 90 ma hollow cathode current. These show larger errors than previous experimental runs at 35 ma. This is believed due primarily to larger and more variable self-reversal effects at the higher tube currents. The assay method is being converted to low current operation and preliminary standards already show greater reproducibility. The working curve for both the 90 ma and the 30 ma current is shown in Figure 3, the upward shift of the 90 ma calibration curve being due to greater self-reversal (intensity reduction) of the Li7 line.

Photoelectric Spectro-Isotopy (G. K. Werner and O. B. Rudolph)

Direct reading photoelectric techniques are being investigated for application to assay problems. Although scanning techniques are simpler to operate, they are in general less accurate than simultaneous line intensity integrations. The optical components necessary for simultaneity of light integration of both isotope spectrum lines consist of slit shifter, cylindrical mirror for magnification, and image splitter. The isotope line separation of only 130 microns on the Rowland circle has been magnified to about 1.5 mm; also, the isotope components have been isolated with the beam splitter and projected on slits of individual phototubes. A converted Kellex amplifier has been used to observe phototube currents, dark currents, and integrated line intensity ratios. Present limitations are in the weak signal response of the phototubes to the infrared.

OPTICAL SPECTROSCOPY

Electrodeless Discharge Sources (W. E. Leyshon and P. M. Griffin)

An experimental vacuum line has been constructed for production of electrodeless discharge tubes containing lithium and its compounds. Several tubes have been made using lithium hydroxide (LiOH), lithium iodide (LiI), and lithium chloride (LiCl) as sample material, and argon and krypton as the carrier gas. Excitation of the lithium spectrum at 1700 megacycles has been negative in all cases.

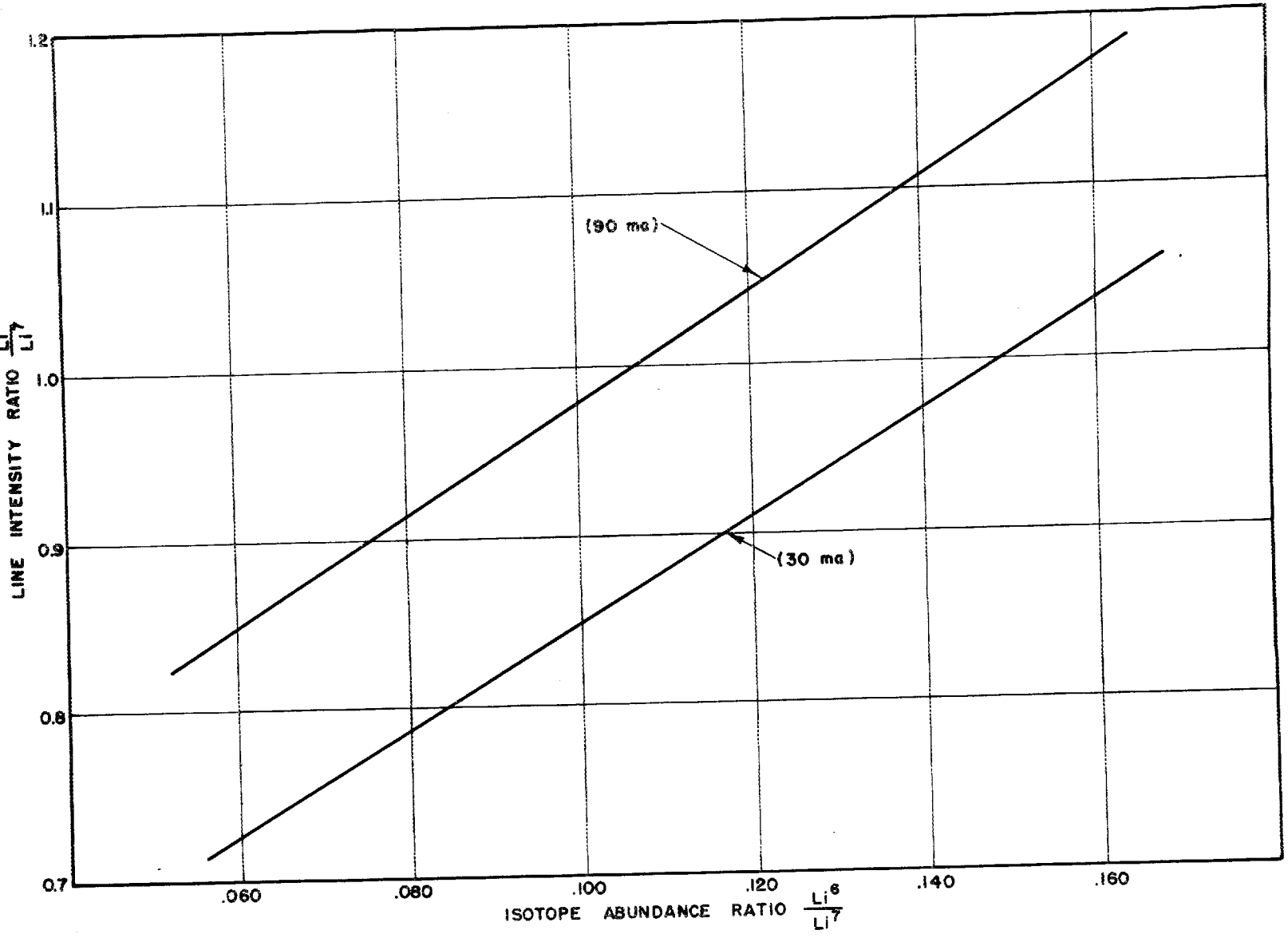

FIGURE 3 CALIBRATION CURVES FOR SPECTRO—ISOTOPIC ASSAY OF LITHIUM

Echelle Spectroscopy (P. M. Griffin)

Echellograms of isotope spectra of uranium were obtained on 40 microgram samples at isotope concentrations of 25, 50, and 75 per cent U235 in U238 (see Figure 4). A plutonium source for handling high level activities is under construction.

Automatic Comparator (G. K. Werner)

A photoelectric comparator scanning and setting device, identical with that designed by Tomkins and Fred of the Argonne Laboratory, has been constructed and is being installed on the SAC (semi-automatic comparator).

SPECTROCHEMISTRY

Trace Contaminants in Alkali Materials (J. A. Norris, M. R. Skidmore, and E. J. Spitzer)

The analysis of alkali materials for trace constituents presents a problem in sensitivity limitation due to the large fraction of low excitation alkali atoms present in the discharge. Prior removal of the alkali has been accomplished chemically and also by vacuum distillation. Calcium may be determined down to 0.7 μg/ml by the copper spark technique with added strontium as the internal standard; vanadium is detectable to 0.5 μg/ml. The present sensitivity limits for other elements in 20 per cent lithium hydroxide ($LiOH \cdot H_2O$) are as follows:

Element	Limit
Cu	0.1 μg/ml
Fe	10
Ni	10
Al	5
Mn	0.4
Cr	1
Mo	2

Miscellaneous (J. A. Norris, Z. Combs, C. E. Pepper, and M. R. Skidmore)

Analyses of mercuric oxide (HgO) matrices have been successfully accomplished to low limits by vacuum sublimation at 400°C of the HgO from the electrode crater. A sensitivity of 0.2 per cent samarium in neodymium

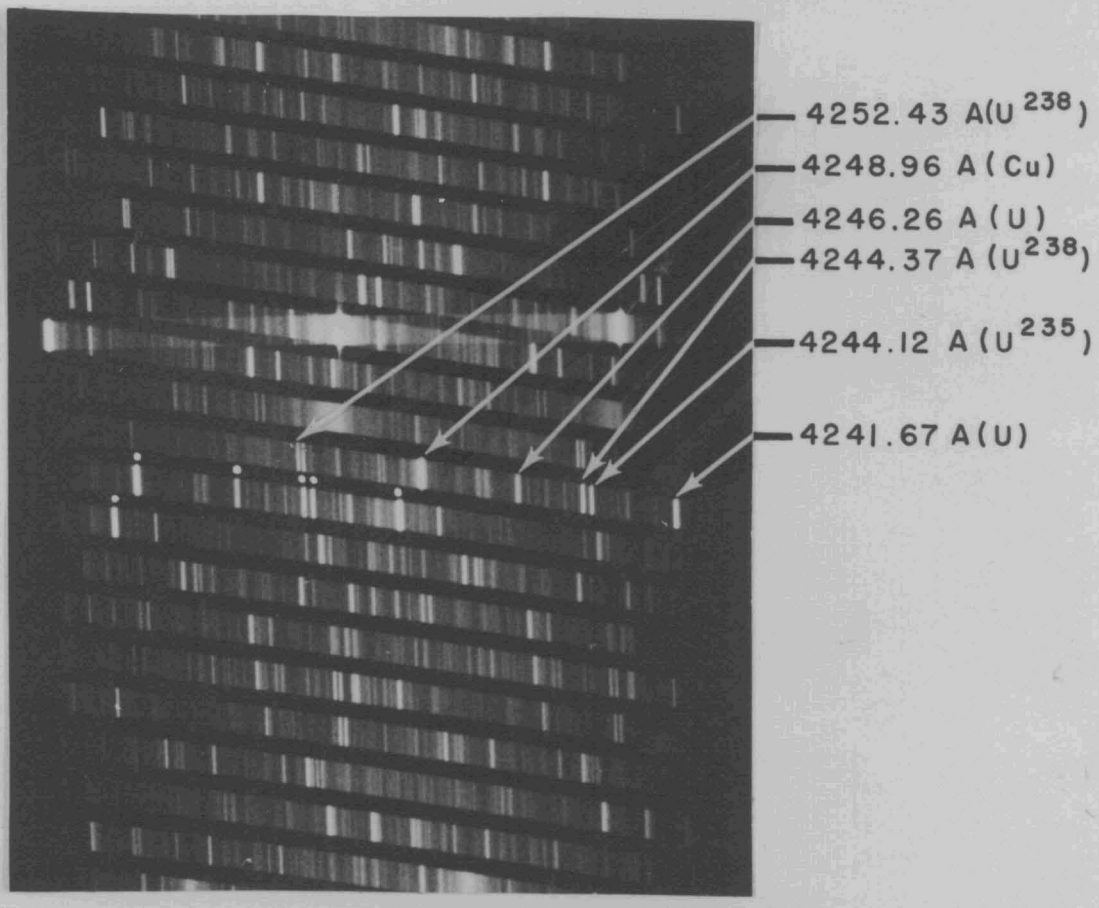

FIGURE 4 ECHELLE SPECTROGRAM OF URANIUM ISOTOPES U^{235} AND U^{238}

in a 1 mg neodymium oxide (Nd_2O_3) sample has been obtained and studies are under way to reduce this to 0.01 per cent samarium. Studies of stainless steels using 5 mg samples burned in direct current arcs show that selective volatilization of tantalum and columbium occurs after iron, chromium and nickel have been removed.

Analyses

A total of 443 samples were analyzed for 6303 element determinations during this quarter.

X-RAY SPECTROSCOPY

X-Ray Spectra of Polonium (W. F. Peed)

X-ray spectra of the L transitions in polonium as obtained with a 25 centimeter Bragg spectrograph have been measured and preliminary data on both K and L spectra are now available. A reporduction of a portion of the L spectrum is given in Figure 5. Construction of the X-ray energy level diagram indicated a previous mis-identification of $\beta 4$ and $\beta 6$. Preliminary results are as follows:

K Spectrum		L Spectrum			
α_2	160.5 XU	ℓ	1280.2 XU	β_1	919.4 XU
α_1	155.6	α_2	1123.7	β_3	905.8
β_1	137.9	α_1	1112.2	γ_1	784.5
β_2	133.0	β_6	965.3	γ_2	769.9
		β_2	927.0	γ_3	762.0

X-Ray Diffraction (H. W. Dunn)

Compound identifications in support of ANP, HRE, and Building 9212 research groups were made in sixty problems. Stainless steels which became magnetic after exposure to hot fluorides showed high concentrations of free iron indicating large scale removal of stainless steel components other than iron by fluoride attack.

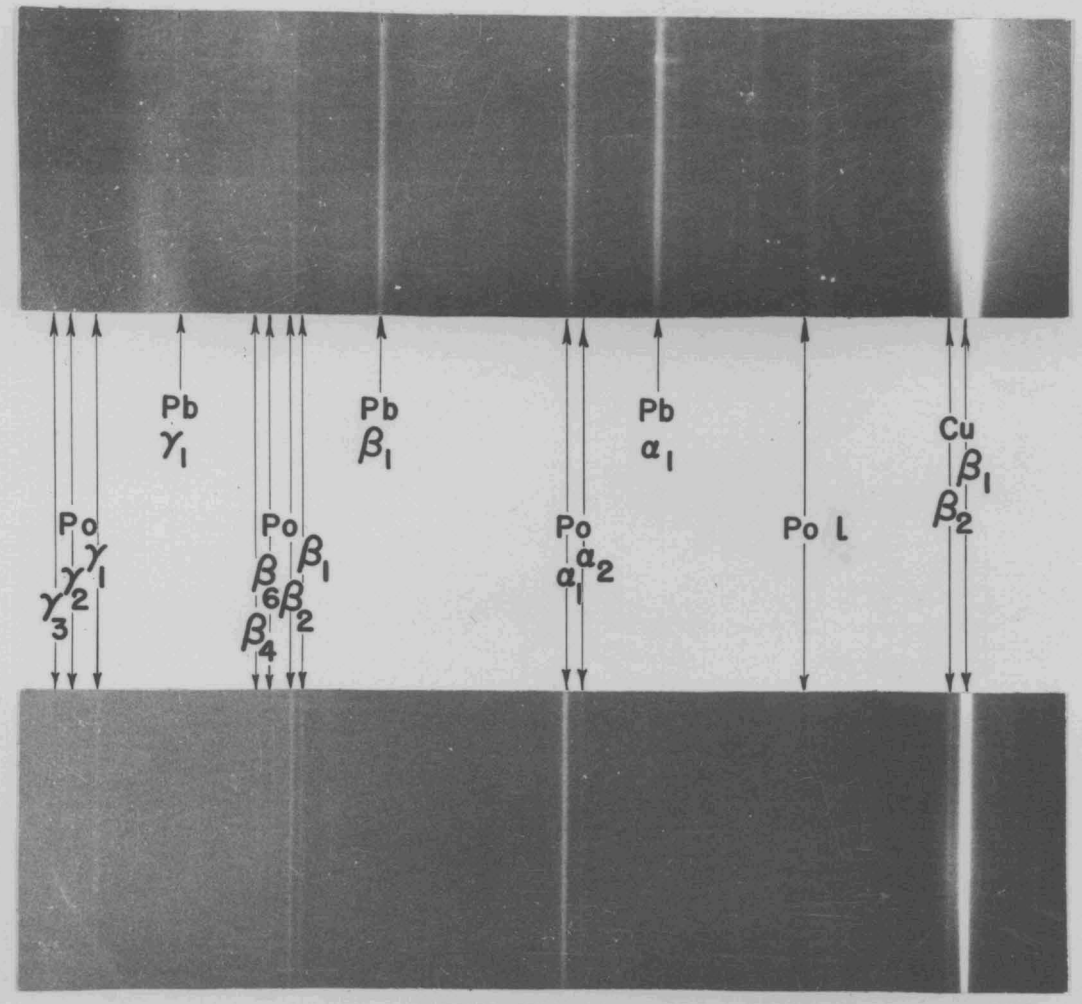

FIGURE 5 L SPECTRUM OF POLONIUM AND LEAD

X-Ray Absorption (H. W. Dunn and W. F. Peed)

The method for determination for uranium in solutions by homochromatic X-ray absorption at the LIII edge of uranium has been investigated further. A series of solutions containing 3686 ppm of uranium and silver (characteristic of elements of atomic number near 47) in concentrations ranging from 14,700 ppm to zero has been investigated. No detectable error was introduced by the silver, although differences as large as 1 to 2 per cent due to instability of the instrument at the time of measurement may have obscured errors of this magnitude.

Counting rate errors have been investigated and correction curves established up to counting rates as high as 40,000 counts per second. At 40,000 input counts, approximately 37 per cent are lost in the present system due principally to amplifier overload.

PUBLICATIONS AND PAPERS

Reports and Papers in Preparation

1. H. E. Walchli, W. E. Leyshon, and F. M. Scheitlin, "The Nuclear Gyromagnetic Ratio of V50, and Measurements on Rb85 and C135."

2. W. F. Peed and H. W. Dunn, "Homochromatic X-Ray Absorption Studies on Uranium in Solutions."

3. J. A. Norris and C. E. Pepper, "Quantitative Spectrochemical Analysis of Rare Earth Mixtures."

4. J. R. McNally, Jr., "Spectro-Isotopy--A New Field of Spectroscopic Endeavor."

5. W. F. Peed, L. E. Burkhart,* R. A. Staniforth,** and L. G. Fauble,** "The X-Ray Spectra of Polonium, Atomic Number 84."

6. G. L. Stukenbroeker and J. R. McNally, Jr., "Isotope Shifts in Thorium for Th230 and Th232."

Reports Submitted for Publication

1. H. E. Walchli, W. E. Leyshon, and F. M. Scheitlin, "The Nuclear Gyromagnetic Ratio of V50 and Measurements on Rb85 and C135," submitted to Physical Review.

2. H. E. Walchli, R. Livingston,*** and William J. Martin,*** "The Nuclear Magnetic Moment of Tc99," submitted to Physical Review.

3. J. R. McNally, Jr., "New Techniques in Mass Analysis," submitted to Bulletin of Applied Spectroscopy.

4. G. K. Werner, "Semi-Automatic Recording of Wavelength Data," submitted to Review of Scientific Instruments.

5. J. R. McNally, Jr., "Atomic Spectroscopy and Separated Isotopes," submitted to American Journal of Physics.

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* Now with Y-12 Plant.

** Mound Laboratory, Miamisburg, Ohio.

*** Chemistry Division, X-10 Area.

6. G. L. Stukenbroeker, D. D. Smith, G. K. Werner, and J. R. McNally, Jr., "A Preliminary Report on Spectro-Isotopic Analysis of Lithium," submitted to Journal of the Optical Society of America.

Reports Published and Papers Presented

1. H. E. Walchli, "The Measurement of Nuclear g-Factors, and the Quantitative Analysis of Isotopes by Nuclear Induction," Symposium on Nuclear Radiofrequency Spectroscopy and Electronic Paramagnetic Resonance, University of Illinois, November (1951).

2. G. L. Stukenbroeker, D. D. Smith, G. K. Werner, and J. R. McNally, Jr., "Mass Assay of Lithium by Optical Spectroscopy," Paper presented to American Institute of Physics Meeting, Chicago, October (1951).

3. D. D. Smith, G. L. Stukenbroeker, and J. R. McNally, Jr., "New Data on Isotope Shifts in Uranium Spectra: U236 and U234," Physical Review 84, 383 (1951).

MASS SPECTROMETER LABORATORY

Russell Baldock, Department Head

INTRODUCTION

During the second quarter of fiscal year 1952 five investigations of aircraft fuels were made, and a sample of technetium oxide was studied.

AIRCRAFT FUEL STUDIES

Uranium Trifluoride (L. O. Gilpatrick and J. R. Sites)

Additional runs were made with the mass spectrometer using very pure uranium trifluoride (UF_3).[2] Using the intensity ratio of the UF_3^+ ion to the UF_4^+ ion, only one of five runs showed any difference from the usual ratio (13) for uranium tetrafluoride (UF_4) material. During this run the ratio remained at 22-23 until, at higher temperatures, near the completion of the run the ratio decreased to 13. Uranium trifluoride dissociates at temperatures necessary to produce a measurable ion beam.[3] A UF_4^+ ion peak was present throughout every uranium ion reception, therefore no pure mass cracking pattern of UF_4 material was obtained. Because of this complication, further attempts to analyze possible radiation damage to UF_4 material by setting up standard mass cracking patterns for the uranium fluorides has been discontinued.

Cyclotron Irradiated Samples (L. O. Gilpatrick and J. R. Sites)

Samples of a three-component fluoride eutectic of sodium fluoride, potassium fluoride (KF), and uranium tetrafluoride (UF_4) were irradiated in the 186-inch cyclotron. These irradiated samples and their corresponding control samples were studies in the mass spectrometer.[4] One typical

2. "Quarterly Progress Report, Aircraft Nuclear Propulsion Project," period ending September 10, 1951. ORNL-1154, page 159.
3. "Quarterly Progress Report, Stable Isotope Research and Production Division, July 1, 1951 to September 30, 1951. Y-821.
4. "Quarterly Progress Report, Aircraft Nuclear Propulsion Project," period ending September 10, 1951. ORNL-1154, page 155.

control sample had a medium amount of silicon tetrafluoride (SiF_4), with a trace of sodium (Na) and UF_4. (See Table II). The corresponding irradiated sample showed a large amount of SiF_4 after a burst in pressure near the melting point of the eutectic (450°C). Above 600°C, a medium amount of UF_4, sodium, and potassium appeared, and the peak at mass 32 greatly increased. Undoubtedly this peak is due to oxygen (O_2^+), but there is a slight possibility that some of the peak might be contributed by sulfur.

Four-Component Fluoride Eutectic (L. O. Gilpatrick and J. R. Sites)

Two samples of the four-component fluoride eutectic of lithium fluoride (LiF), sodium fluoride (NaF), potassium fluoride (KF), and uranium tetrafluoride (UF_4) were examined.[5] The first sample was prepared by the Materials Chemistry Division (MCD) of the Y-12 plant by mixing and heating the components to form the eutectic, and then grinding the cooled mass to a powder. The second sample was a dry mixture of the specified proportions of the fluorides from the bulk chemicals used by the Aircraft Nuclear Propulsion (ANP) Division. A small portion of this mixture was heated in the source oven inside the mass spectrometer to form the eutectic.

The first of these samples showed a small amount of mercury which disappeared when the temperature was increased, and a medium amount of SiF_4 appeared above the melting point of the eutectic. There was a trace of sodium and potassium, but no uranium or lithium peaks appeared. Above 600°C a considerable amount of lead chloride ($PbCl_2$) of unknown origin showed up. The second sample also had a medium amount of SiF_4 and potassium, a small amount of sodium, but no uranium or lithium peaks. The mass 32 peak increased considerably at the higher temperatures.

Above 460°C the sodium fluoride material alone gave a moderate Na^+ ion peak with no other noteworthy features. Potassium fluoride gave a medium K^+ ion peak above 630°C with a small amount of SiF_4 at this high temperature. Prominent unidentified peaks appeared with this material, a pair of peaks at masses 97 and 99, and an isolated peak at mass 76 that rose and declined in intensity apparently independent of any other peaks.

The temperature of the lithium fluoride material alone was raised to 730°C, but no lithium peaks were found. A small amount of hydrogen chloride (HCl) and sodium appeared, and a very small amount of SiF_4. At the higher temperatures the mass 32 peak intensity increased over a factor of three.

5. "Quarterly Progress Report, Aircraft Nuclear Propulsion Project," period ending December 10, 1951. ORNL-1170.

TABLE II

MASS SPECTROMETER STUDIES OF AIRCRAFT FUELS

Mass	Identity	UF_3	Fuel				Fuel Components			
			Three Component		Four Component					
			Control	Irradiated	MCD	ANP	NaF	KF	LiF	UF_4
23	Na	o	t	h	t	t	h	o	t	o
32	O_2,S	d	p	u	p	u	p	d	u-i	u
36	HCl	1	o	t	1	1	o	1	1	1
39	K	t	o	h	h	i	o	h	o	o
49	BF_2^+	m	m	m	m	m	o	m	m	m
78	?	o	o	o	t	o	m	m	m	m
85	SiF_3^+	m	m	i	m	m	m	m	t	m
97	?	o	t	o	o	m	o	m	o	o
202	Hg	o	o	1-t	1	o	o	1-t	o	o
208	Pb	o	o	o	i	o	o	o	o	o
238	U	h(UF_4)	t	h	o	o	o	o	o	h(UF_5)

p Present during entire run
1 Present at low temperature
m Present at medium temperatures
h Present at high temperature
o None reported
t Trace amount
i High intensity
u Intensity increases at high temperatures
d Intensity decreases at high temperatures

The UF_4 material alone had a strong persistent UF_5 pattern which changed to the normal UF_4 pattern at the higher temperatures during the latter part of the run.

In the fluoride runs, the intensity of the carbon dioxide (CO_2^+) peak at mass 44 went through a rather high maximum in the 350°-400°C temperature range, indicating the presence of dissociating carbonates. These runs were also characterized by the presence of boron trifluoride (BF_3), as indicated by the BF_2^+ ion peaks at masses 48 and 49, and by a small pair of peaks at 10 and 11. A trace of HCl appeared in several of the runs. The prominence of the 97-99 pair of masses in the KF run may have been due to K_2F^+ ions. No NaF^+, KF^+, or LiF^+ ions were observed in any run where they might have appeared.

Silicon Tetrafluoride Observations (L. O. Gilpatrick and J. R. Sites)

Even though platinum charge material containers were used, SiF_4 still persisted as a major factor in the fluoride runs. This strongly indicated that silicon was a contaminant in the constituent chemicals, or was introduced during preparation. In the event of very intense SiF_4 peaks, measurable amounts of metastable mass fragments from the most intense SiF_3^+ ion are formed. The singly ionized, doubly ionized, and the metastable mass cracking patterns of SiF_4 are presented in Figure 6.

Sublimation Studies (L. O. Gilpatrick)

A preliminary sublimation study of the fluoride eutectic components was started, and some standard UF_4 was prepared by removing the UF_5 found in commercial UF_4 by vacuum decomposition.

FISSION PRODUCT STUDIES

Technetium (L. O. Gilpatrick and J. R. Sites)

A 20 mg sample of technetium in the form of ammonium pertechnicate (NH_4TcO_4) was obtained from Dr. G. W. Parker of X-10 who separated the material from fission products.[6] This sample was converted to an oxide by carefully heating with H_2O_2 and NH_4OH. The mass cracking pattern indicated

6. Parker, G. W., James Reed, and J. W. Ruch, "Isolation of Milligram Amounts of Element 43 From Uranium Fission." AECD-2043.

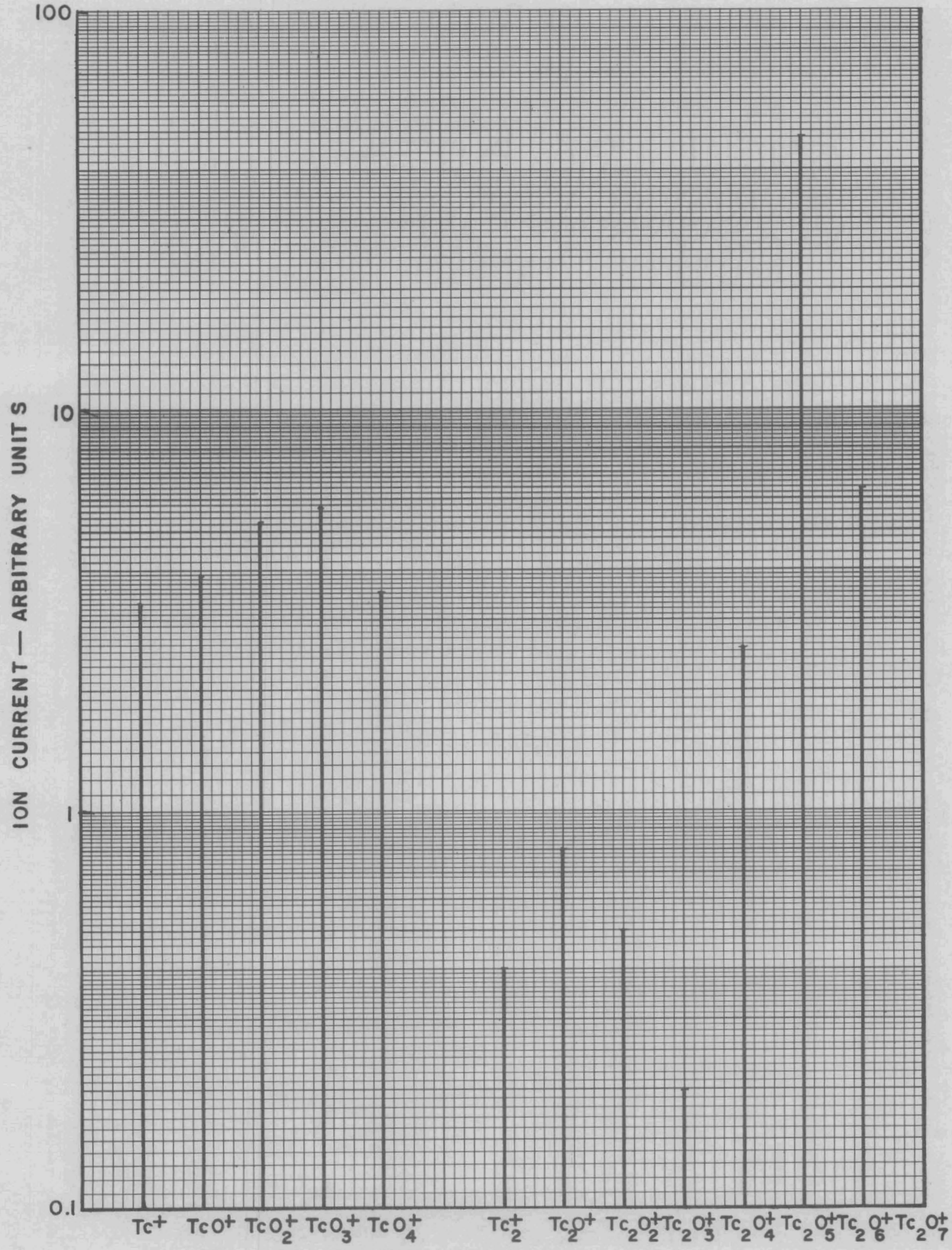

FIGURE 6 MASS CRACKING PATTERN OF TECHNETIUM OXIDE

a complex oxide structure, or mixture, as shown in Figure 7. Only technetium of mass 99 was observed. Upper limits on other possible isotopes, with respect to Tc99, were set in a previous investigation as 1/30.[7] Values set from the data of this study are: Tc97, 1/3,000; Tc100, 1/5,000; Tc101, 1/5,000. The parent materials were technetium tetroxide (TcO_4), and technetium heptoxide (Tc_2O_7), with indications of NH_4TcO_4 and of some acid ions such as $HTcO_2$. The corresponding oxides of rhenium were also found. This element was added as a carrier in the fission product separation because its chemical behavior is very similar to that of technetium. In addition, some ions of selenium appeared: Se^+, SeO^+, SeO_2^+, and Se_2^+.

The appearance potential curve for Tc99 is plotted with the appearance potential curve of argon for this instrument in Figure 8. The onset value for argon is within one-half volt of the optical spectroscopy ionization potential of 15.76 volts. However, any accurate onset value for technetium is lost in the long tailing off of the curve. The appearance potential from this data does not remotely approach the predicted optical spectroscopy ionization potential of about 7.5 volts. The curve presented can only indicate that the Tc99 ions are due to fragmentation of more complex ions, and not by ionization of technetium atoms. With increased sensitivity of the mass spectrometer electrometer the tail of the technetium curve might be followed to much lower electron voltages, and a more comparable appearance potential determined.

7. Inghram, M. C., David C. Hess, Jr., and Richard J. Hayden, "Mass Spectrometric Study of Fission Element 43," Physical Review 72:1269, (1947).

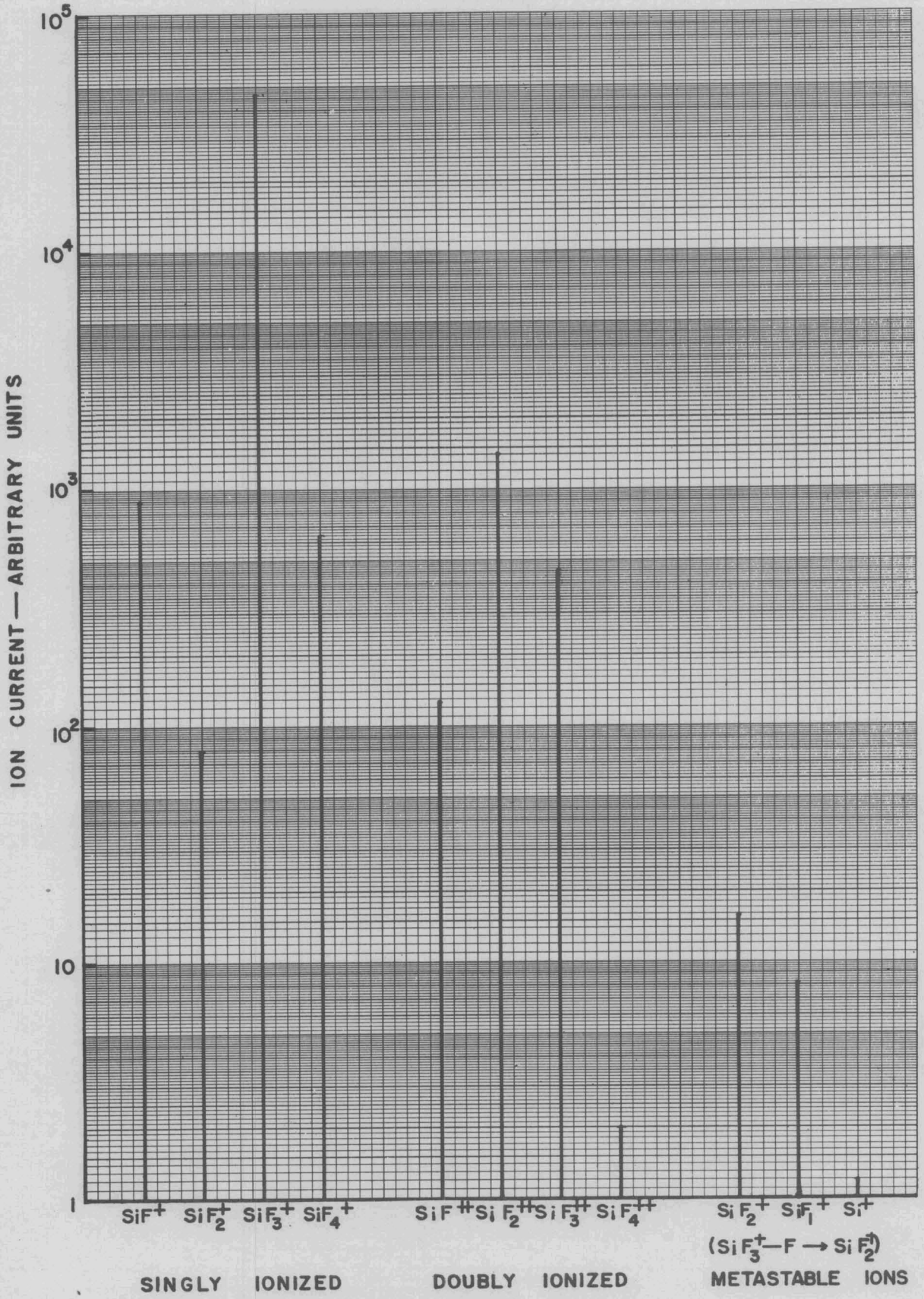

FIGURE 7 MASS CRACKING PATTERNS OF SiF_4 FROM FLUORIDE EUTECTIC

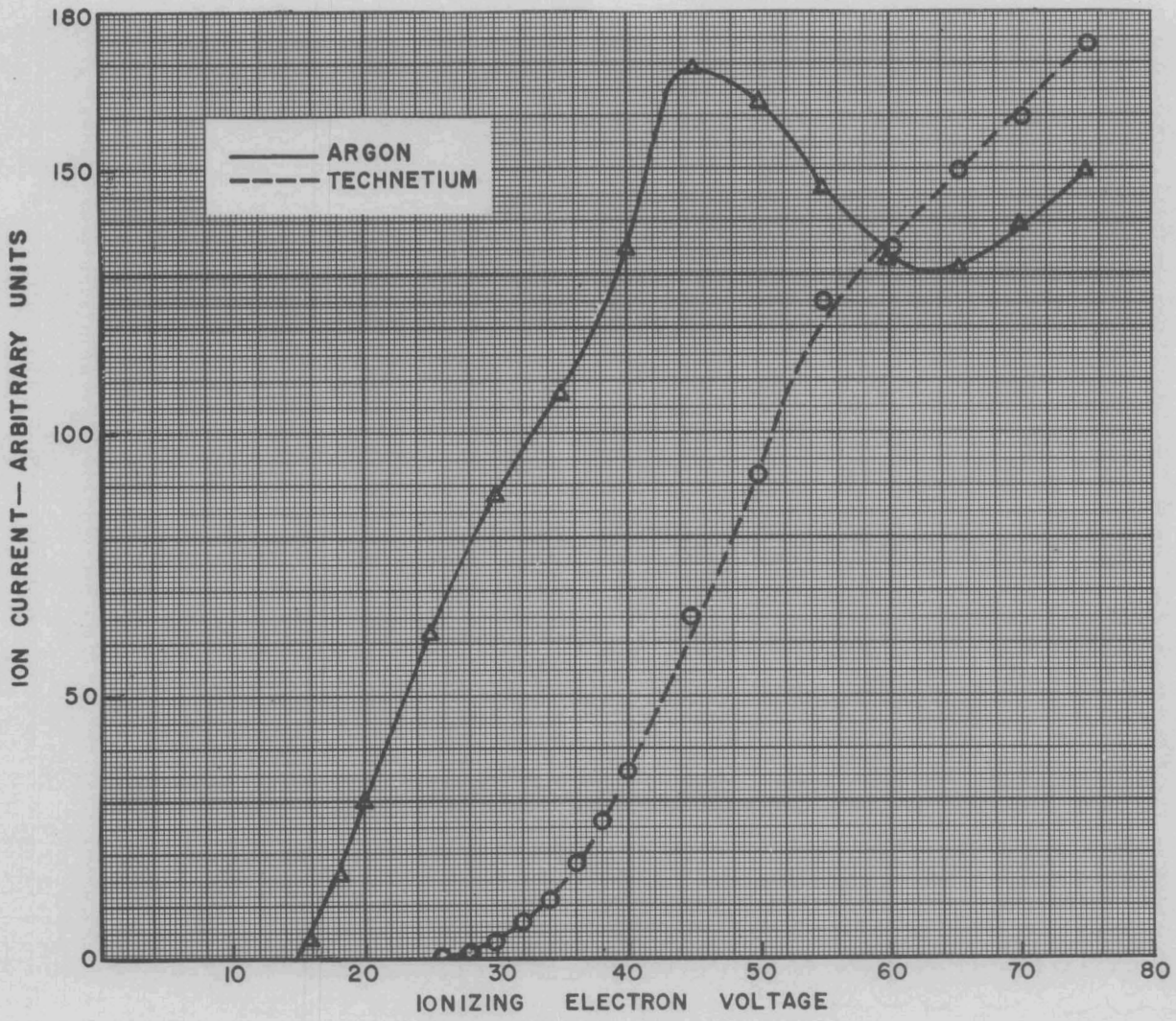

FIGURE 8 APPEARANCE POTENTIAL CURVES OF TECHNETIUM AND ARGON