**Isotope Separations Section Progress
Report for Quarter Ending March 31, 1976**

E. Newman

OAK RIDGE NATIONAL LABORATORY

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CHEMICAL TECHNOLOGY DIVISION

ISOTOPE SEPARATIONS SECTION PROGRESS REPORT

FOR QUARTER ENDING MARCH 31, 1976

E. Newman

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ISOTOPE SEPARATIONS SECTION PROGRESS REPORT
FOR QUARTER ENDING MARCH 31, 1976

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SUMMARY

The activities of the Isotope Separations Section of the Chemical Technology Division are described for the quarter ending March 31, 1976. Included is the status of the current electromagnetic separation of the isotopes of chlorine, tin, and ytterbium. The research and development activities on the 255° separators to improve the isotopic purity of germanium isotopes are also discussed. An improved chemical recovery-recycle system for ytterbium charge material and a study of the factors involved in recovering ^{202}Hg from oil-coated collector plates are also described. The preparation of thin osmium targets of platinum, and samarium isotopes by ion deposition in the sector isotope separator has been achieved. Summary tabulations are given of elements chemically processed and new lots of isotopes released to inventory.

STABLE ISOTOPE SEPARATIONS

Isotopic separations of tin and ytterbium were continued throughout the quarter, and a chlorine separation that was initiated early in January also ran throughout the period. These separations were conducted in a total of twenty-four 180° separators on a 24-hr, 5-day-week basis. The ytterbium separation, which was started in late March 1975, with the goal of obtaining ~6 g of $\geq 20\%$ ^{168}Yb , was terminated on March 31, 1976, after attaining an estimated quantity equivalent to the stated requirement. A new short separation of germanium isotopes is scheduled to begin in the 255° units on April 1, in order to recover ≥ 10 g of $\geq 90\%$ ^{73}Ge . This material, which is of a higher assay quality than previously attained, is intended for Sales distribution in uses such as Mössbauer research.

DEVELOPMENT ACTIVITIES

 ^{255}O Separator Studies

Since the last report, studies of the ^{255}O separation system have been continued with a dual objective: first, to acquire a better understanding of the basic factors limiting separator performance, and, second, to determine ^{255}O separator parameters that would yield $\geq 90\%$ ^{73}Ge at a reasonable ion output (20 to 25 mA Ge^+). Both objectives were pursued simultaneously. In the interest of economy in the forthcoming separation, an early decision was made to limit investigations to the use of existing 12-in.-radius ion-exit-slit geometry. Similarly, the choice of charge material was reduced to GeCl_4 , based simply on its availability by a specified utilization date. Germanium tetrachloride was known to present neutral-vapor problems that resulted in the dilution of germanium isotopes with normal germanium. Thus the present work incorporated calcium-pumping as a means of controlling this problem and included a receiver having masses of ^{76}Ge and ^{72}Ge which were electrically monitorable. This receiver design (1) permitted periodic monitoring of the ^{72}Ge - ^{73}Ge valley current for subsequent use in determining the quality of beam focus attained and (2) provided a means for the estimation of the quantity of isotopic material being collected.

Comparison of the performance of the systems having ion exit slits with dimensions of $1/8 \times 3$ in. and $3/16 \times 3$ in. showed essentially equal quality of beams, with total ion output to the receiver averaging 19 and 27 mA, respectively. Both ion output and beam quality (peak-to-valley ratio) were shown to be pressure dependent, as indicated in Fig. 1. The curves plotted are typical of responses obtained by optimizing system parameters and then throttling the pumping system in steps to produce an increase in system pressure. System pressure was attributable to GeCl_4 input (~ 1 g/hr), atmospheric inleakage (~ 0.05 g/hr), and calcium vaporized onto the tank walls (~ 1.5 g/hr). The results leave little doubt that the effective separation of germanium isotopes requires operation at low pressure to overcome three basic difficulties: (1) low ion output,

(2) poor beam quality or focus, and (3) contamination by deposition of neutral vapors of germanium.

In trial runs that were designed to test proposed germanium-component geometry, 14 unit start-ups were introduced; the ion output averaged 26.0 mA for 60 hr and the peak-to-valley ratios averaged 90 to 1. Recovered ^{73}Ge assayed 92.2%, indicating that the system and the equipment chosen are capable of providing the $\geq 90\%$ ^{73}Ge set as a goal in the forthcoming separations series.

Fig. 1. Germanium ion output and beam quality as a function of tank pressure.

Ytterbium Recycle Purification

During the current ytterbium isotopic separation, which ran for ~1 year in order to obtain needed ¹⁶⁸Yb, it became economically necessary to recover and recycle the un-ionized portion of the charge material. At the outset, typical precipitation-type chemical separations were attempted to separate a large quantity of impurities (principally copper and iron) introduced by washing separator units. These procedures, however, became extremely time-consuming and inefficient due to the large volumes of solutions encountered.

As a means of reducing chemistry time and, at the same time, providing a purer charge material, an established solvent extraction procedure using pure tri-N-butyl phosphate (TBP) was tried. As a consequence, it was found that chemistry time could be reduced by about 15 hrs with very little (~0.002%) ytterbium lost, and would result in ytterbium oxide charge material of higher purity.

Basically, the principle is rather simple, although a large-volume evaporator and mixer-settler extraction equipment are necessary. Nitric acid wash solutions of calutron components (mainly sources and liners) are filtered on paper to remove solids, and the filtrates are evaporated to concentrate ytterbium. The solids (largely carbon) are ignited and the ash leached with hot HNO₃ (~10 M). The insolubles are centrifuged for clarification, and the centrifugate is evaporated in a tantalum-lined evaporator using 30-psi steam pressure.

Approximately 8 liters of filtered evaporator concentrate is adjusted to ~10 M in HNO₃ and is then extracted batchwise with approximately 10 liters of pure TBP. Each batch of concentrate contains from 100 to 700 g of ytterbium. An additional 4 liters of TBP is run through the mixer-settler extractor to ensure ytterbium removal. Ytterbium is stripped from the TBP with distilled water and recovered by precipitation with oxalic acid. After ignition, ytterbium oxide is pressed into buttons and recycled in the separator by means of in situ chlorination with carbon tetrachloride.

Mercury-202 Retention Study

For some time now, the technique for collecting mercury isotopes in calutron units has been based on the decomposition of films of Octoil-S [di(2-ethylhexyl)sebacate] either in water-cooled copper pockets or on water-cooled copper plates. The collection principle appears to be dependent on the adsorption of mercury on the decomposed oil films. This procedure, which is a compromise between earlier trapping systems that employed water-cooled silver collector pockets and expensive liner refrigeration, produces relatively pure mercury isotopes without the need for a refrigeration system. Retention, that is, the ratio of monitored beam to the material actually recovered, is quite variable in mercury collections due to the high vapor pressure of the impinging metal ions. Retention of mercury (on oil films, in particular) appears to be dependent on the vapor pressure of mercury, ion output, size of collection plate, quality of the film itself, and the saturation of the deposit.

After the recovery and refinement of the products from a recent separation, a correlation of the ^{202}Hg retention values was made. The basis for the correlation was in terms of ion output and quantity monitored onto a copper plate, with an effective collection area of approximately 18 in.². In the separation series that was terminated on December 31, 1975, 15 runs were made in which the estimated weight of ^{202}Hg was <300 mg and 27 runs in which the estimated weight was >300 mg. The runs that had an estimated weight of <300 mg of ^{202}Hg had an average product retention of 40.5%, while those with an estimated weight of >300 mg of ^{202}Hg had an average retention of 28.1%.

The correlation of retention values, based on 10 runs of highest retention and 10 runs of lowest retention, along with parameters such as beam collection time (innage hours) and collection rate, is shown in Table 1.

Table 1. Correlation of ²⁰²Hg retention values

	Innage (hr)	Rate (mg/ hr)	Estimated weight (g)	Recovery weight (g)	Retention (%)	Assay (%)
Average for 10 runs with highest retention	70.0	4.22	0.295	0.144	48.8	95.6
Average for 10 runs with lowest retention	92.7	4.26	0.395	0.073	18.5	96.4

These values indicate that, after ~300 mg of isotopic mercury has been deposited on this particular surface area, the surface becomes saturated, and additional mercury ions are lost from the collector. It seems evident, therefore, that it would be economically advantageous in future separations which utilize the same collection system to use a combination of parameters (natural abundance, ion output, and reception time) that would lead to only 300 mg of deposit.

TARGET PREPARATION BY ION DEPOSITION

Isotopically enriched thin targets of ¹⁸⁶Os, ¹⁹²Pt, ¹⁹⁴Pt, and ¹⁵⁰Sm were prepared during the quarter in the 180°-sector isotope separator (ORSIS) by the surface deposition method. Two targets of each isotope were prepared with deposition thicknesses of 25 µg/cm² for ¹⁹⁴Pt and ¹⁵⁰Sm and 50 µg/cm² for ¹⁸⁶Os and ¹⁹²Pt. The substrate, in every case, consisted of 75 µg/cm² carbon foil. The ion beam was decelerated from about 35 kV to ~200 eV at the point of impact on the substrate. The isotopic purity of the wanted isotope was estimated to be >99% in each case, and the initial feed was normal isotopic material. All of the targets were prepared for use in coulomb excitation studies.

CHEMICAL PROCESSING

Material in Process

The following elements were in chemical processing during the quarter: chlorine, mercury, thallium, tin, and ytterbium.

Material Added to Inventory

Table 2 identifies the isotopic material that was released or made available to Sales during this report period; the appropriate separation data are also included. In addition to the eight new lots of isotopes released to Sales, 19 loan-return samples were processed and returned to Research Materials Collection (RMC) for inventory.

Table 2. Operations status for the period January-March 1976

Isotope	Series	Series Schedule		Hours		Charge material	Recovery (g)	Purity (%)	Requirements	
		Start	Finish	Innage	Tank		Actual	Actual	Grams	Purity (%)
⁵⁶ Fe	OQ	6/23/69	2/26/71	61,231	85,128	FeCl ₂	8916.66	99.87	6000	>99.8
¹⁹⁶ Hg	QB	8/11/75	12/31/75	3,619	4,704	Hg	0.255	20.99	50	>48
²⁰² Hg	QB	8/11/75	12/31/75	3,619	4,704	Hg	4.487	96.32	50	≥95
¹²³ Te	PS	3/15/74	3/21/75	22,817	27,340	Te	1.092	46.85	13	>80
¹²⁴ Te	PS	3/15/74	3/21/75	22,817	27,340	Te	26.932	95.42	100	>95
¹²⁴ Te	PS	3/15/74	3/21/75	22,817	27,340	Te	26.545	93.75		
¹²⁵ Te	PS	3/15/74	3/21/75	22,817	27,340	Te	29.814	95.67	100	90-97
²⁰³ Tl	QA	8/6/75	12/31/75	11,969	14,304	Tl	524.022	92.36	600	>90

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