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SUMMARY

Isotope Separation and Purification

Helium-3 As a part of the effort to establish the vapor pressure of helium-3 as the international temperature scale between 0.3° and 3.2°K, the manometric corrections from helium-4 to helium-3 are being determined. The results of a single run did not agree with the predicted values.

Neon Isotope Separation Calculations indicate that a two to one thermal diffusion column cascade is more efficient than a three to one cascade.

Gaseous impurities in neon were reduced from 3.40 to 0.26% by passing the impure gas through a charcoal trap at liquid nitrogen temperature.

Krypton Isotope Separation About 15 liters of gas containing 9% krypton-78 and krypton-80 were accumulated. Calculations to determine the shape factors for the thermal diffusion system were completed.

Carbon-13 A six-stage cascade system was converted to an eight-stage cascade system of ten hot-wire and concentric-tube columns. Prior to the conversion the six-stage system enriched natural methane to 74 per cent. Additionally, 74 per cent of that enriched methane product was mass 17. The concentric-tube columns completed the purification of a commercially enriched methane from 60 per cent to over 90 per cent carbon-13 in the total carbon. About eight grams of the methane product have been accumulated.

Alpha and Neutron Sources

A method has been developed for fabricating threshold detectors by sealing them in copper cans. Twenty-two plutonium threshold detectors, returned because of high wipe counts, were removed from the aluminum cans; then the detectors were sealed inside copper cans by tungsten inert gas welding. The recanned threshold detectors had zero wipe counts.

Radioelements

Residue Adsorption The erratic behavior of barium-lanthanum-140 was traced to a macroscopic impurity in the stock solution. A separation of this impurity by cation exchange indicated tentatively that it is stable strontium.

Determination of Coincidence Correction The multiple source method for determining coincidence correction was compared with the double source method. The more difficult multiple source method offered no advantages and had the distinct disadvantages of complexity and increased working time in comparison to the double source method.

Polonium Research The radioactive decay of polonium-208 and polonium-209 is being studied in detail. A gamma ray at 565 kev in the polonium-208 spectrum may actually be two or more gamma rays arising from different spin states in the daughter nucleus, bismuth-208. This premise was confirmed by a detailed study of the electron conversion spectrum of the polonium-208-polonium-209 mixture. Conversion electrons apparently arise from gamma energies at 535 ± 10 , 565 ± 10 , and 595 ± 10 kev.

The visible absorption spectrum of polonium(IV) in hydrochloric acid was measured with a mixed isotopic source of polonium-208 and polonium-209. An attempt to measure the visible absorption spectrum of polonium(II) in hydrochloric acid was unsuccessful.

The half-life of polonium-209 was determined in several different experiments. The most reliable data, based on a measurement of total polonium by absorption spectrophotometry and total alpha activity by gamma counting, indicate that the half-life of polonium-209 is 36 years. The currently accepted half-life value is 103 years.

The half-life of polonium-210 is being determined to the nearest minute at a 95 per cent confidence level using gadolinium polonide.

Promethium Separation and Purification A study is being conducted on the separation of neodymium and samarium in preparation for the isolation of promethium. The study of a method for separating promethium from americium is nearly complete. Americium exists in promethium samples at a concentration of about three atom per cent.

ISOTOPE SEPARATION AND PURIFICATION

Processes are being developed for separating and purifying the isotopes of the noble gases and carbon. Potential sources of supply are being evaluated.

HELIUM-3 THERMOMETRY

In cooperation with Los Alamos Scientific Laboratory and the National Bureau of Standards, Mound Laboratory is attempting to establish the vapor pressure of helium-3 as the international temperature scale between 0.3° and 3.2°K. As a part of this effort the manometric corrections in this temperature range must be established.

A run was made on the experimental vapor pressure thermometer using helium-4 instead of helium-3. The liquid helium-4 in the bath was evacuated to a pressure of 3.2 torr, which corresponds to a temperature of 1.48°K. A *Texas Instrument* bourdon tube was used to detect any thermomolecular pressure difference between the 1/4-inch and 1/32-inch diameter pressure sensing tubes. At 3.2 torr as measured at the warm end, a difference of 0.12 torr was predicted from calculations using the Weber Equation. [This was confirmed experimentally by Roberts and Sydoriak.]¹

Having operated the system successfully with helium-4, a run was made using helium-3, but because the bath temperature could not be controlled with sufficient accuracy, only one measurement was made. At a bulb temperature of 1.48°K the barytron gage indicated a pressure difference of 14 microns between the 1/32-inch and 1/4-inch tubes. This correction is too low for helium-4 when a pressure correction of 120 microns is predicted.

NEON ISOTOPE SEPARATION

Thermal Diffusion The flows in the four-column, two-stage neon hot wire thermal diffusion unit were adjusted to enrich neon-20 to about 99.7 per cent at a flow rate of approximately 315 milliliters per hour, and neon-22 to about 99.7 per cent at a flow rate of approximately 30 milliliters per hour. The intermediate stream contained about three per cent neon-21 at a flow rate of 16 milliliters per hour.

The performance of a two to one cascade arrangement was computed and compared with the three to one overlap arrangement presently in use. The concentrations and flows for feed points at three locations along the cascade are shown in Table 1. The best feed point (F, in Figure 1) is the point along the column which gives an enrichment of 99.9 per cent for both neon-20 and neon-22. The product rate of neon-22 is 37 cc per hour, which is larger than the present rate of 30 cc per hour. The separation of both isotopes is aided by the additional 12 feet of column for the enrichment of neon-22, not available in the three-one overlap arrangement.

A study was also made of a 2-1-1 cascade. The results are given in Table 2. As shown in Figure 2, F₁ is a good feed point for this cascade and the corresponding concentrations and flows are reasonable.

The profile of the present three to one overlap arrangement was calculated and compared to the experimental profile taken from sample ports on columns three and four. The results are given in Figure 3.

A significant amount of remixing occurred in the second stage, which was caused by about 70 per cent neon-22 from the first stage being fed into the second at a point in the column where the concentration was measured to be about 90 per cent. The feed point was moved four feet up the column which should give the situation

¹T. R. Roberts and S. G. Sydoriak *Thermomolecular Pressure Ratios for Helium-3 and Helium-4* *Phys. Rev.*, 102, 304 (1956).

Table 1

**DATA FOR THE 2-1 CASCADE FROM
THE CASCADE COMPUTER PROGRAM**

Feed Point	Flow (ml/hr)			Concentration (%)	
	Feed	Product	Waste	Top Ne 20	Bottom Ne 22
F ₁	956	53	903	96.29	99.9
F ₂	559	47	512	99.17	99.9
F ₃	407	37	370	99.87	99.9
F ₄					
F ₁	600	50	550	99.16	99.9
F ₂	Pressure Too High				
F ₃	To Obtain Separation				
F ₄	Of Neon 22 To 99.9%

Table 2

**DATA FOR THE 2-1-1 CASCADE OBTAINED
FROM THE CASCADE COMPUTER PROGRAM**

Feed Point	Flow, ml/hr			Concentration (%)		$\frac{K_c}{K_d}$
	Feed	Product	Waste	X _w Ne 20	X _p Ne 22	
F ₁	2220	123	2097	96.12	99.90	10
F ₂	1688	116	1572	97.5	99.90	10
F ₃	1307	104	1203	98.69	99.90	10
F ₄	989	87	902	99.49	99.90	10
F ₅	790	63	627	99.87	99.90	10

Figure 1. Two-one Column Arrangement

of $C_F = C_O$ in the second stage where C_F is the feed concentration and C_O the concentration in the column at the feed point. This approaches the ideal thermal diffusion column and should supply more accurate data to compare with theoretical calculations and to lead to improved performance of the columns.

Neon Purification The gaseous impurities (H_2 , He, H_2O , N_2 , O_2 , etc) were removed from a batch of neon by passing the gas through a charcoal trap at liquid nitrogen temperature ($-195.8^\circ C$). The gas composition before and after the run was determined. After the run, the charcoal trap was warmed to $23^\circ C$, and the gas which evolved (about 2.1 liters) was sampled. Finally, the cold trap was warmed to about $350^\circ C$ to expel the remaining adsorbed gases (about 2 milliliters). With one pass through the charcoal trap the total impurities were reduced from 3.40 per cent to 0.26 % in 8.6 liters of neon.

KRYPTON ISOTOPE SEPARATION

A two-column hot-wire thermal diffusion cascade unit with an infinite reservoir at the bottom processed natural krypton in the first step to obtain 90% krypton-78 and 80. Batch runs were made starting with 2.7% krypton-78 and 80 at a column pressure of 330 torr. Overhead product was withdrawn at a rate of 15 milliliters per hour when a concentration of about 12% krypton-78 and 80 was reached. As the process continued, the concentration decreased to about eight per cent. Withdrawal of material reduced the column pressure to about 290 torr. About 15 liters of gas containing approximately 9% krypton-78 and 80 were accumulated.

Calculations for this process used shape factors based on the Lennard-Jones (12-6) potential and transport coefficients, H , K_C , and K_d , defined by McInteer and Reisfeld.² The transport coefficient, H , describes the radial transport of the desired isotope produced by the thermal diffusion effect; K_C and K_d describe, respectively, the remixing which occurs due to convection currents and to ordinary diffusion along the column. The pressures at the maximum separation factor where $K_C = K_d$, and the pressure at 90 per cent maximum separative work, where $K_C = 10 K_d$, were considered.

The performance of the column was described by the differential equation from Jones and Furry

$$r = H_c (1-c) - (K_C + K_d) (dc/dz)$$

to give the net transport rate of the desired isotope in grams per second.

²McInteer, B. B., and Reisfeld, M. J., J. Chem. Physics, 33:2, 571, (August, 1960).

Figure 2. Two-one-one Arrangement

XENON ISOTOPE SEPARATION AND PURIFICATION

An eight-column, four-stage hot wire thermal diffusion unit for depleting fission product xenon of xenon-131 was modified to a swing arrangement between columns in cascade. The use of 1/8-inch intercolumn piping showed a restricted flow when the top of the first stage reached a concentration of 17 per cent xenon-131, with the infinite feed reservoir (at a much lower concentration) connected to the top of first stage. The column piping has been changed to 3/16 inch. The swing flow was set at about 90 milliliters per cycle on a four-minute cycle. A perceptible pressure change was noted in the feed reservoir, indicating transport between the reservoir and the first stage.

CARBON ISOTOPE SEPARATION

The objectives of the carbon-13 program are the preparation of gram quantities of 90 per cent carbon-13, the evaluation of carbon monoxide and methane as feed materials for thermal diffusion column separation, and the evaluation of chemical exchange, mass filtration, and distillation to enrich carbon-13.

Thermal Diffusion A six-stage cascade system of six hot wire columns and two concentric tube columns for the separation of carbon-13 from natural methane reached a mass-17 enrichment of 74% in 74% methane. The system was then increased to 10 columns by adding two 1-1/4 x 3/4-inch concentric tube columns in series between the last two stages. The gas used to fill the added columns was partially enriched methane obtained from previous thermal diffusion experiments.

When sufficient material of 80% mass-17 enrichment became available, an attempt was made to determine the extent of contamination of the mass-17 ($C^{13}H_4$) fraction with monodeuterated carbon-12 methane ($C^{12}H_3D$). If no chemical exchange took place in the thermal diffusion columns, the natural abundance ratio of $C^{13}H_4$ to $C^{12}H_3D$ should prevail, and 5.3 per cent of the mass-17 in all parts of the cascade should be $C^{12}H_3D$.

A special sample bottle containing a platinum wire heater was prepared. The bottle was filled with methane from the column system. The methane was subsequently partially decomposed into carbon and hydrogen by heating the platinum wire. The gaseous sample was submitted for mass spectrometric analysis. After analysis, the sample bottle was evacuated and refilled with oxygen.

The carbon in the bottle was burned to carbon dioxide by heating the bottle with a torch. A mass spectrometric analysis was made of this material. The results are given below:

Mass-17 enrichment of methane in sample	68.53%
Deuterium content of hydrogen from sample	0.39%
$C^{12}H_3D$ content of methane	1.55%
$C^{13}H_4$ content of methane	66.98%
$C^{13}H_4$ content of methane from analysis of CO_2	

The results of the hydrogen analysis indicate that exchange took place, since only 2.3 per cent of the mass-17 was found to be $C^{12}H_3D$. It is also possible that excess hydrogen was in the sample from outgassing of the platinum wire. This would make the measured $C^{12}H_3D$ content too low. The procedure will be checked for this possibility.

Production of 90 per cent enriched $C^{13}H_4$ from enriched feed was stopped. All available feed material has been processed to yield 16.2 liters of methane which contains 8.54 g of C^{13} .

THERMAL DIFFUSION RESEARCH

Low Temperature Thermal Diffusion Factor The thermal diffusion factor is important for theoretical calculations because of its sensitive dependence on the intermolecular potential function. Because of increasing quantum effects below room temperature, its value in this range is of special interest. The thermal diffusion factor over the temperature range of 10 to 250°K is being studied using a two-bulb apparatus described previously (MLM-1177).

The calculation of the correction for the gas pumped out of the interconnecting tube after each expansion during a run has been completed and the results are shown in Table 3. For these calculations, the convergence criterion on the material balance was set so that it must agree to within 0.1 per cent for each expansion. The results show that the only correction occurred at 100°K. At 10°K, there is no correction in the composition change because the thermal diffusion coefficient, α is 0.011. At 200°K, there is no correction because the ratio of the temperature difference to the average temperature is small. At 100°K, both of these factors combine in such a way so as to require a large correction.

Table 3

CALCULATED COMPOSITION FOR TWO-BULB THERMAL DIFFUSION EXPERIMENT

Mean Temperature (°K)	Temperature Difference (°K)	Final Composition ^a		Percentage Correction to Composition Change by "Exact Method"
		Light Isotope	Heavy Isotope	
10	6	51.709	48.290	0
	4	51.123	48.876	0
	2	50.546	49.453	0
100	20	48.483	51.516	97.2
	15	48.742	51.257	78.7
	10	48.995	51.004	50.0
	5	49.228	50.771	0
200	20	48.634	51.365	0
	15	48.965	51.034	0
	10	49.297	50.702	0
	5	49.629	50.370	0

^aConcentrations are given in percentages after 20 expansions.

MASS FILTER

The mass filter is a device similar to an electromagnetic separator, but it employs the action of ions in a rotating electrostatic field to attain mass dispersion. This device is attractive because it avoids many of the secondary effects that limit the usefulness of other electromagnetic and electrostatic methods. Also, precise electrostatic fields are easier to generate and control than magnetic fields.

The device consists of an ion source, four rods or electrodes, and an ion collector enclosed in a vacuum system. A rotating field is generated by RF power applied to the quadrupole electrode system. Under certain conditions of accelerating potential and frequency of rotation of the field, ions of a very narrow mass range can be focused on the collector.

A multipole system was constructed for measuring the capacitance of a parallel array of 25 mass filters. The rods are removable so that they can be arranged in an array of 2 x 2 rods to 6 x 6 rods as shown below:

Figure 3 Arrangement of Mass Filter Rods

Aluminum rods 7/16-inch in diameter were used with Lucite insulators and spacers. Capacitance measurements, made with a Ballantine capacitance meter, indicate that the total capacitance of 25 mass filters in such an array is approximately one-half the total capacitance of 25 individual mass filters.

Since the power requirements of a mass filter are directly proportional to its capacitance, a parallel array of mass filters would be more efficient than the same number of mass filters built as independent units. Construction of such an array would, of course, be more complex than that of a single unit, but should be less costly than construction of an identical number of single units.

ALPHA AND NEUTRON SOURCES

The radioisotope distribution activity at Mound Laboratory provides for the sale of radioisotopes and for related services. As part of this activity Mound Laboratory supplies alpha or neutron sources which cannot be supplied by industry. The radioisotopes polonium-210 and plutonium-239 are used in the fabrication of these sources. In addition, extensive research and development is being conducted to develop new techniques for manufacturing these sources.

NEUTRON CALIBRATION

A survey was made of different types of neutron spectrometer systems to select and build a system for use at Mound Laboratory. Characteristics of an ideal spectrometer system include:

- a. Low sensitivity to background
- b. Good energy resolution
- c. High efficiency over a large range of energies
- d. Rapid accumulation and reduction of data

These optimum characteristics are approached in varying degrees by different types of spectrometer systems. The best choice will be that system which most closely approaches the characteristics of an ideal system and is useable for continuous neutron spectra of radioactive sources in the energy range from 0 to 14 Mev.

The system which has been selected as best meeting the requirements actually consists of two different types of spectrometers. A spectrometer containing a single stilbene crystal will be used for the energy range above 1 Mev and another spectrometer containing a set of polyethylene spheres will be used below this range.

Briefly, the operation of the stilbene crystal spectrometer is described below:

Neutrons incident upon the crystal are elastically scattered by protons within the crystal. The energies of the protons are determined from the scintillations produced within the crystal and amplified by a photomultiplier. Gamma ray background is eliminated by a simple circuit which utilizes differences in pulse shapes of scintillations produced by electrons and protons. Differentiation of the measured proton spectrum yields a neutron spectrum, which is corrected for wall effects and second scattering to give the incident neutron spectrum.

The operation of the polyethylene spheres is described below:

Five spherical pieces of polyethylene of different diameters, each with a small hole into which a Li^6I crystal is inserted as a neutron detector are used. The sensitivity of each sphere is a function of the neutron energy. The relative counting rates for the different sized spheres will be dependent upon the energy of the source of neutrons. With five different spheres four data points for a spectrum are obtained. If a spectrum is known above a particular neutron energy, E_n , the four data points may then be spaced in the interval below E_n .

The principal advantage of the stilbene crystal spectrometer over other systems is the relatively rapid accumulation and reduction of data. The rate of accumulating data is up to one order of magnitude faster than the next most efficient electronic spectrometer operated under conditions of comparable resolution. Other systems, e.g., proton recoil telescope and emulsions, are capable of better resolution, but this is achieved only by a much greater expenditure of total time.

POLONIUM SOURCES

The four windowed sources described previously (MLM-1177) were checked periodically. There appears to be no difference between those with the window in contact with the polonium and those with spaced windows.

Two additional gold plated sources were fabricated using a backing of platinum foil brazed to stainless steel. These, in addition to the two sources with gold backing, appear to be retaining the major portion of their polonium.

PLUTONIUM SOURCES

A method was developed for welding threshold detectors in copper cans using TIG welding. Twenty-two plutonium threshold detectors, returned because of high wipe counts, were recanned in copper containers. Checks of the outer container revealed that the plutonium was completely incarcerated.

RADIOELEMENTS

Basic and applied research is being conducted on a number of radioelements to determine their physical properties, to develop analytical techniques, and to study the basic radiochemistry involved. Of particular interest are alpha emitters, their decay chains, their isotopes, and their chemical homologs.

RESIDUE ADSORPTION

Considerable difficulty has been experienced in purifying the barium-lanthanum-140 received from Oak Ridge. While the material appears to be radiochemically pure, it contains up to 3.5 milligrams per milliliter of non-volatile solids, as indicated by the analysis accompanying each shipment. While this amount of impurity does not seriously affect the use of the barium-lanthanum-140 as a radioactive tracer, it represents a significant interference in studies requiring carrier-free source material.

In an effort to isolate the barium-140 in a carrier-free condition, three successive cation exchange separations were made on the most recent shipment received. The barium-140 was evaporated to dryness, made one normal in nitric acid, and loaded on a small cation exchange column (Dowex-50W-X8; 200-400 mesh). The column was washed repeatedly with small portions of one normal nitric acid until barium-140 appeared in the effluent. The eluent was changed to three normal nitric acid, and the effluent was collected in 0.25-milliliter fractions. The two central fractions, containing approximately 90 per cent of the barium-140, were combined, evaporated to dryness, and inspected visually. A white residue indicated that the barium-140 still contained a macroscopic impurity. All other fractions were discarded, and the central barium-140 fraction was again loaded on a Dowex-50W column in one normal nitric acid. The same procedure was repeated and the central fraction was again retained, while the head and tail fractions were discarded.

A third fractionation was made on a fresh cation exchange column. The head fraction, containing the early barium-140 breakthrough, had a fairly heavy residue on drying. The tail fraction, which showed early lanthanum-140 breakthrough, had only a slight residue, as did the central fraction, containing approximately 85 per cent of the barium-140 remaining after three cation exchange fractionations.

The white residue in the head fraction was readily soluble in water. The solution was made one normal in nitric acid and transferred to a platinum disk, where it was dried and converted to a hydroxide with ammonium hydroxide. The hydroxide residue was leached with distilled water and the solution was transferred to a second platinum disk. Gamma spectrometry showed that approximately 25 per cent of the lanthanum-140 was transferred with approximately 90 per cent of the barium-140.

The macroscopic appearance of the residues on the platinum disks was significant; the residue on the first platinum disk, containing 75 per cent of the lanthanum-140, was homogeneously spread over the entire platinum surface, but the residue on the second platinum disk dried in a ring at the periphery of the open platinum surface. Furthermore, within five minutes after the transferred solution had been dried at 90°C., it was again wet with moisture absorbed from the air.

The ion exchange behavior of the impurity in the head fraction and the deliquescence of its hydroxide residue lead to the tentative conclusion that the impurity is stable strontium, which was probably added as a carrier during the purification of the barium-140 prior to shipment.

Although there are numerous methods reported for the quantitative separation of barium and strontium, all of them involve the addition of undesirable non-volatile carriers or complexing agents. It seems likely, therefore, that the best purification procedure for the present purpose will be that already adopted, that is, the repeated elution of strontium and barium from a cation exchanger with nitric acid, visual inspection of the residues from the various fractions, and recycling of the central barium fractions until a weightless residue is obtained.

DETERMINATION OF COINCIDENCE CORRECTION

Probably the most commonly used procedure for determining the dead time and coincidence correction of a counting apparatus is the double source method. This method consists, in its essentials, of counting two radioactive sources separately and simultaneously; because of dead time losses, the sum of the counting rates for the individual samples is always greater than the observed counting rate of the combined samples.

This method is inherently inaccurate, because the usual computation is based on an approximation which is invalid at higher counting rates. In an earlier report (MLM-1117, page 6), an alternate method of computation was proposed, which eliminates this approximation:

$$r = \frac{\frac{a}{n_{12}} - \frac{1}{n_1}}{a - 1} ; \quad (1)$$

$$a = 1 + \left[\frac{\frac{1}{n_1} - \frac{1}{n_{12}}}{\frac{1}{n_2} - \frac{1}{n_{12}}} \right]^{1/2} \quad (1a)$$

where r is the dead time of the counting instrument, n_1 and n_2 are the observed counting rates of the individual sources, and n_{12} is the observed counting rate of the combined sources. The factor, a , is numerically equal to the ratio between N_{12} , the true counting rate of the combined sources and N_1 , the true counting rate of the first source.

While this method of computation eliminates the customary approximation used in the double source method, it does not resolve the problem of selecting the proper function to describe the counting losses. Kohman³ rejects the assumption that the dead time can be treated as a constant in the equation

$$N = \frac{n}{1 - nr} \quad (2)$$

upon which the double source method is based. Instead, he proposes that the calculation be based on a power series with arbitrary constants,

$$N = n + an^2 + bn^3 + cn^4 + \dots, \quad (3)$$

which can be evaluated from a series of double source determinations by the method of least squares, provided the number of determinations is greater than the number of constants to be evaluated.

To test the Kohman procedure and, at the same time, to determine the function which best describes the dead time of a proportional counter, 17 double source determinations were made. The data were inserted in equation (4), and solved for 2, 3, 4, 5, and 10 arbitrary constants.

$$n_1 + n_2 - n_{12} = a(n_{12}^2 - n_1^2 - n_2^2) + b(n_{12}^3 - n_1^3 - n_2^3) + c(n_{12}^4 - n_1^4 - n_2^4) \quad (4)$$

For comparison, the data were also inserted in equations (1), which represent the case of a single constant, r , and assume that the arbitrary constants of Equations (3) and (4) are integral powers of r . The results of

³Kohman, T. P., Natl. Nuclear Energy Ser., Div. IV - Plutonium Project Record, vol. 14B, page 1655 (McGraw-Hill, N. Y.) 1949.

these computations are summarized in Table 4. Since the agreement between the computed values of $(N_1 + N_2)$ and N_{12} is an indication of the validity of the computation, the computed value of N_{12} is shown in Table 4 above the sum of the computed values of N_1 and N_2 .

Table 4
COMPARISON OF DOUBLE SOURCE AND MULTIPLE SOURCE METHODS
FOR COMPUTING COINCIDENCE CORRECTION

Observed (cts./min.)	Computed Counting Rate (cts./min.)*					
	1	2	3	4	5	10
99,951	101,278	100,886	101,435	100,501	101,057	101,260
$N_1 + N_2$	101,278	101,073	101,369	100,847	101,171	101,277
99,956	101,272	100,891	101,440	100,506	101,062	101,265
$N_1 + N_2$	101,271	101,072	101,368	100,846	101,170	101,276
100,170	101,183	101,110	101,660	100,723	101,281	101,485
$N_1 + N_2$	101,184	101,138	101,434	100,911	101,235	101,342
540,144	577,588	572,855	581,350	570,403	575,678	578,173
$N_1 + N_2$	577,588	574,005	580,715	570,664	575,922	578,346
541,894	579,411	574,839	583,366	572,385	577,677	580,178
$N_1 + N_2$	579,412	575,898	582,643	572,546	577,825	580,260
542,194	577,524	575,179	583,711	572,725	578,020	580,521
$N_1 + N_2$	577,525	575,132	581,863	571,785	577,055	579,486
543,256	580,234	576,384	584,935	573,929	579,233	581,739
$N_1 + N_2$	580,233	577,075	583,842	573,717	579,009	581,451
543,434	583,856	576,586	585,140	574,130	579,436	581,943
$N_1 + N_2$	583,858	578,968	585,769	575,599	580,912	583,365
543,709	581,260	576,898	585,457	574,442	579,751	582,258
$N_1 + N_2$	581,261	577,841	584,622	574,479	579,779	582,226
785,833	870,932	861,427	873,503	858,378	866,211	869,751
$N_1 + N_2$	870,932	862,565	874,459	858,538	866,366	870,082
787,694	875,068	863,695	875,791	860,635	868,490	872,041
$N_1 + N_2$	875,068	865,635	877,586	861,491	869,453	873,187
787,855	870,573	863,891	875,989	860,830	868,687	872,239
$N_1 + N_2$	870,574	863,770	875,683	859,642	867,580	871,299
787,972	870,852	864,034	876,133	860,972	868,831	872,383
$N_1 + N_2$	870,853	863,971	875,888	859,844	867,783	871,503
1,015,831	1,166,363	1,152,094	1,166,273	1,146,362	1,157,266	1,162,660
$N_1 + N_2$	1,166,361	1,152,078	1,169,169	1,147,167	1,157,771	1,162,780
1,395,617	1,709,142	1,683,806	1,704,806	1,673,558	1,688,730	1,695,542
$N_1 + N_2$	1,709,148	1,680,196	1,703,950	1,674,296	1,689,542	1,696,412
1,396,254	1,706,624	1,684,758	1,705,781	1,674,519	1,689,697	1,696,510
$N_1 + N_2$	1,706,618	1,679,418	1,703,164	1,673,521	1,688,759	1,695,625
1,573,368	2,013,777	1,958,075	1,988,373	1,956,046	1,974,263	1,982,789
$N_1 + N_2$	2,013,768	1,963,804	1,989,982	1,956,136	1,974,303	1,982,793

*For two, three, four, five, or ten arbitrary constants by the method of least squares, and for one constant by Equations (1).

It is immediately apparent from Table 4 that Equations (1) yield nearly perfect agreement between N_{11} and $(N_1 + N_2)$, showing that the customary approximation of the double source method has been eliminated; such differences as occur are entirely attributable to rounding-off errors in the computation.

It is also apparent that the agreement between comparable values for the computations made with different numbers of arbitrary constants is not particularly good. If it is assumed that the case of 10 constants is inherently more valid than those of fewer constants in the least squares solution, then the double source method, with a single constant, gives at least as good agreement as does the multiple source method with 2, 3, 4, or 5 constants.

It is noteworthy that, as the number of constants in the least squares solution increases, the computed counting rate for a given observation oscillates around the counting rate computed for 10 constants. The reason for this behavior can be seen in Table 5, which lists the values of the constants found for each least squares solution of the multiple source data. It is seen that each constant changes not only in magnitude, but in sign, so that the arbitrary decision to solve for a given number of constants affects the computed counting rate according to the sign and magnitude of the first constant eliminated.

Table 5

COMPARISON OF CONSTANTS FOUND IN THE LEAST SQUARES
SOLUTION OF THE MULTIPLE SOURCE METHOD

Constant	Number of Constants Sought				
	2	3	4	5	10
a	$+0.8949 \times 10^{-7}$	$+0.1518 \times 10^{-6}$	$+0.3779 \times 10^{-7}$	$+0.1114 \times 10^{-6}$	$+0.1328 \times 10^{-2}$
b	$+0.4189 \times 10^{-13}$	-0.3505×10^{-13}	$+0.1878 \times 10^{-12}$	-0.1703×10^{-13}	-0.1916×10^{-5}
c	-	$+0.2868 \times 10^{-19}$	-0.1476×10^{-18}	$+0.1145 \times 10^{-18}$	$+0.1340 \times 10^{-7}$
d	-	-	$+0.4794 \times 10^{-25}$	-0.1061×10^{-24}	$+0.9352 \times 10^{-10}$
e	-	-	-	$+0.3380 \times 10^{-31}$	$+0.8936 \times 10^{-11}$
f	-	-	-	-	-0.1525×10^{-12}
g	-	-	-	-	$+0.8758 \times 10^{-15}$
h	-	-	-	-	-0.2580×10^{-17}
i	-	-	-	-	$+0.8557 \times 10^{-20}$
j	-	-	-	-	-0.2093×10^{-22}

For the same reason, Table 5 fails to give any clue to the form of the function which best describes the coincidence losses in a half-life determination.

It must be concluded that the multiple source method offers no advantages over the double source method for computing dead-time corrections, and introduces distinct practical disadvantages because of the arduous computations and the loss of significance characteristic of the method of least squares.

POLONIUM RESEARCH

Polonium-208 and Polonium-209 The mode of decay of polonium-208 and polonium-209 is being studied. The structure of the peak in the polonium-208 spectrum at 565 kev indicates the presence of two or more

gamma rays. The resolution by NaI(Tl) scintillation detector for this peak is, for example, about half as good as it should be. (The peak referred to here is that obtained in coincidence with the 295 kev photon and is free of interference from the 900 kev gamma ray in polonium-209.) It was assumed that this peak is actually a multiplet; thus, a series of coincidence measurements was made as a function of the angle between the two 3 x 3-inch NaI(Tl) detectors.

Presumably, the various gamma rays making up this peak arise from various spin states in bismuth-208, the daughter nucleus of polonium-208. The multipolarity of the emitted radiation will differ and the angular distribution of the radiation patterns will vary for the various states. Some change of peak shape with angle might be expected.

Figure 4 shows the result of this study. Although the peak value itself remains constant with angle, there are shifts and changes in the leading and trailing edges in keeping with the above description. These curves are normalized at the peak.

The existence of several close lying states in the vicinity of 565 kev leads to the probable existence of one or more low-lying levels in bismuth-208 (below 100 kev) in keeping with theoretical calculations.⁴ A series of triple coincidence measurements was made utilizing two amplitude restrictions. Gates were set at about 50 to 90 kev on each of the two gate detectors to detect the x-radiation or low energy gamma rays associated with a given decay mode in each case.

A typical result obtained in this study is shown in Figure 5. The three peaks at 295, 565, and 900 kev normally obtained in gamma-gamma coincidences with gating on the x-ray peak were observed; however, the peak ratios changed radically.

Many competing processes are involved, thus the interpretation of the results in Figure 7 is difficult. If grating conditions were satisfied by an x-ray and low energy gamma ray the ratio of the 295 and 565 kev peaks would have been substantially larger. Since this was not the case, it has been tentatively concluded that the gate conditions were satisfied predominantly by: (1) the x-ray resulting from electron capture, and (2) the x-ray resulting from internal conversion of the 295 kev state. The peak at 860-890 kev is attributed to chance coincidences from polonium-209 decay.

Additional studies were made with a thick depletion depth gold-silicon surface barrier detector on the conversion electron spectrum of polonium-208-polonium-209. These data are being analyzed. It appears, however, that three gamma rays contribute to the 565 kev peak in bismuth-208. The most exacting series of energy measurements indicate that the energies of the gamma rays giving rise to these conversion electron peaks are 535 ± 10 kev, 565 ± 10 kev, and 595 ± 10 kev. This is in keeping with results mentioned above on gamma-gamma coincidences as a function of angle between detectors (Figure 5).

Macrochemistry of Polonium Several radiochemically pure, carrier-free sources of mixed polonium-208 and polonium-209 were prepared. Millicurie quantities of polonium were purified from the Monel fittings on which it had been deposited by the following technique:

1. The Monel parts were rinsed with 3 M HCl. In most cases this mild treatment removed 90% of the polonium activity in three rinses. In some cases the Monel pieces were completely dissolved in aqua regia, which was subsequently de-nitrated with formic acid before proceeding.
2. The solution, containing nickel, copper and polonium, was passed through a bed of powdered bismuth metal, on which the polonium was deposited by spontaneous electrodeposition. The precious metals, selenium, tellurium, copper, mercury, and several other metals below bismuth in the electromotive series were also deposited in this step.

⁴S. Wahlborn, *Levels in Bismuth-206*, Nuclear Physics, 3, 644-48 (1957).

Figure 4. Coincidence Spectrum of 565 keV Peak as Determined by the Angular Orientation of Three Detectors.
(gate set on 295 keV peak; resolving time 45 microsecond)

Figure 5. Triple Coincidence Spectrum of $Po^{208} Po^{209}$
(gates 1 and 2 set at 50-90 keV, 2780 minute count)

3. The bismuth metal powder was then dissolved in aqua regia, and denitrated with formic acid. Stannous chloride was then added, precipitating polonium, tellurium, selenium, copper, silver, gold, mercury, and probably iridium.
4. The precipitate was dissolved in a mixture of 3 M HCl and 30% H₂O₂. Tellurium and selenium were precipitated as the metals with hydrazine dihydrochloride. At this point, the pink color of polonium(II) in solution became visible. The excess hydrazine was destroyed with HNO₃, and the solution was treated with concentrated ammonia, precipitating polonium hydroxide, and leaving the impurity elements in solution as ammonia complexes.

Experiments described in the section entitled **Polonium-209 Half-Life** (page 20) showed that a considerable amount of impurity was present in one polonium sample prepared in this manner. The impurity elements have not been identified.

Visible absorption spectra of polonium(IV) in 1.5 molar hydrochloric acid were obtained from 400 to 650 millimicrons ($m\mu$), and are shown in Figure 8. The spectra were measured with a Coleman Model 14 Universal Spectrophotometer, using two-dram glass vials. The reference solution was 1.5 molar hydrochloric acid. The cell light path was estimated to be 1.2 cm. No effort was made to use matched sample and reference cuvettes, because of the severe alpha contamination which would result. The strong absorption band at approximately 425 $m\mu$ agrees with the position of the band observed by Hunt³ in hydrochloric acid solutions of polonium-210.

It appears unlikely that any impurity contributed to the visible absorption spectra in the two samples. One of the samples was prepared from a dissolved Monel fitting; the other sample was purified from the first rinse of a hot piece of Monel. The contribution of any impurity element would be much greater in the first case; in fact, the optical density of the samples at 425 $m\mu$ was directly proportional to the total alpha activity of the samples, indicating that the absorption was due only to polonium(IV).

Figure 6. Optical Absorption Spectra of Po(IV) in 1.5 M HCl.

³D. J. Hunt, *Absorbancy Studies of Polonium Complexes in Chloride Solutions*, MLM-979, 1954.

An attempt to measure the absorption spectrum of polonium(II) in hydrochloric acid was not successful, because the concentration of polonium(II) was not high enough to cause significant absorption. The slightly pink solution was stable for a period of several weeks, indicating that measurements will be possible with larger amounts of polonium in solution. (Po^{210} can be reduced to pink polonium(II) in HCl, but oxidation due to alpha radiolysis is so rapid that a measurement of the visible absorption spectrum is not possible.)

Polonium-209 Half-Life Several experimental approaches are being employed to determine the half-life of polonium-209. They are: (1) measuring the polonium-208-polonium-209 mass ratio in a sample in which the polonium-208-polonium-209 alpha decay ratio is known; (2) weighing and counting a sample containing polonium-208 and polonium-209; (3) counting a sample containing polonium-208 and polonium-209 and determining the polonium content by analytical chemistry techniques; and (4) counting samples containing polonium-208, polonium-209 and polonium-210 with high-resolution alpha pulse-height analysis equipment.

Three runs were made in the experiment in which the mixed polonium isotopes were weighed and counted. The polonium was plated from 1.5 molar nitric acid on platinum microfoils at an electrode potential of 0.0 volts (relative to a saturated calomel electrode). The microfoils were weighed on a quartz-fiber microbalance, and total alpha activity on the microfoils was determined in a low-geometry alpha counter (LOGAC), and by gamma counting. Because of the low counting rates obtained in the LOGAC, counting statistics were poor; more accurate results were obtained from gamma counting. Three plating runs, each with duplicate samples, were made, using approximately 30 millicuries of polonium.

In the first experiment a significant quantity of an impurity was present in the polonium. The impurity was electroplated under the experimental conditions. The material which plated on the microfoils was black, in contrast with the dull gray appearance of polonium-210 under similar conditions. The half life of polonium-209 was calculated to be 282.1 years, which is an order of magnitude higher than expected. The error is due to the weight of inert material.

The second plating experiment was carried out under identical conditions to the first experiment. It was expected that the first plating had preferentially removed the impurity so that only polonium would be plated. This proved not to be the case: the polonium-209 half-life was calculated to be 327.8 years.

The third plating was performed under the same conditions as the first two experiments. Once again, the preferential removal of polonium rather than the impurity was demonstrated. The calculated half-life increased to 349.7 years.

The plated microfoils and the depleted plating solution are being reworked to remove the platinum and the impurity element. Further experiments will be performed when the polonium has been purified.

Two auxiliary experiments were required to calculate the half-life of polonium-209 from the plating experiment data. The alpha counting ratio of polonium-208-polonium-209 in the isotopic mixture was determined by alpha pulse-height analysis to be 17.80 ± 0.05 . The alpha/gamma counting ratio was measured by counting several samples of the polonium-208-polonium-209 mixture plated on silver foils on a 3 x 3 inch NaI(Tl) crystal, and in a low geometry alpha counter. The alpha/gamma counting ratio was found to be $(6.17 \pm 0.05) \times 10^3$ alpha disintegrations per gamma count.

In the third experimental method, the total polonium concentration was determined from optical density measurements, which were described in *Macrochemistry of Polonium*, Hunt (loc. cit.), showed that the polonium(IV) absorption band at $420 \text{ m}\mu$ obeyed Beer's Law; that is, the polonium(IV) concentration is proportional to the optical density. Hunt gave the absorbancy index as $(1.06 \pm 0.03) \times 10^4$ for the $420 \text{ m}\mu$ band. Using this value, the polonium concentrations could be calculated from the optical density and the cell light path. Total alpha activity was determined by gamma counting 10 microliter aliquots of both solutions. The experimental data and the calculated results are given in Table 6. Agreement between the two samples is

good, considering the experimental approximations used. The largest experimental error is in the determination of the cell light path. The glass vials employed in the measurements of optical densities were not matched, and the vial diameters were measured with a ruler. Further work, in which matched light cell will be used, is now in progress.

In the counting experiments, the counting rates of polonium-208, polonium-209, and polonium-210 as a function of time, are compared. Since the half life of polonium-210 is known with high precision, it should be possible to determine the polonium-208 and polonium-209 half-lives. One series of counts has been made. No decay of polonium-209 has been observed, within experimental error, over a period of 363 days.

Table 6
EXPERIMENTAL RESULTS OF Po^{209} HALF-LIFE DETERMINATION

Measurement	Sample 316,033-1	Sample 316,056-1
Optical density, 425 $m\mu$	0.420	0.611
Po(IV) concentration, M	3.302×10^{-5}	4.803×10^{-5}
Total Po weight, grams	24.10×10^{-6}	35.04×10^{-6}
Total alpha activity, d/m	1.869×10^{10}	2.908×10^{10}
Po^{209} alpha activity, d/m	9.889×10^8	1.539×10^9
Po^{208} alpha activity, d/m	1.770×10^{10}	2.754×10^{10}
Po^{208} weight, grams*	13.39×10^{-6}	20.84×10^{-6}
Po^{209} weight, grams	9.71×10^{-6}	14.20×10^{-6}
Po^{209} half life, years	37.27	35.04

* Po^{208} half life was taken to be 1055 days.

Polonium-210 Half-Life Sample A polonium-210 sample was prepared to determine the half-life to the nearest minute level at a 95% confidence. This error would be a factor of nine less than the error in previous determinations which gave a result of 138.3749 ± 0.0050 days. The improvement is expected for three reasons: (1) a more accurate calorimeter apparatus; (2) additional measurement for statistical control and (3) less polonium-208 and polonium-209 in the sample.*

The outside dimensions of the sample are 0.281 O.D. by 0.438 inch long which permits the sample to be measured in both microcalorimeters 60 and 67 when the source has decayed to about one or two curies. As of noon November 6, 1963, the power of the source was 3.1266 watts (97.61 curies) which is the average of three measurements in Calorimeter 90. The polonium in the sample was reacted with gadolinium and contained in two stainless steel containers, each sealed by welding. It is not expected that the half-life measurements will be affected by using a polonium compound.

An error of one minute out of 138.375 days is 0.0005 per cent. At first, it is surprising that such a small error in the half-life can come from measurements accurate to about 0.01 per cent. The primary reason for this is due to the nature of an exponential curve when extended many half-lives. Given two power measurements of W_1 and W_2 watts with a known error for each and separated by Δt days, the error in the half-life can be computed by Equation 1.

*R. C. Lange and K. C. Jordan Po^{208} and Po^{209} In Reactor-Produced Po^{210} Nuclear Science and Engineering (in press).

$$\text{Error in } T_{1/2} = \frac{\Delta t \ln 2}{\ln^2 \frac{W_1}{W_2}} \sqrt{\left(\frac{\text{Error in } W_1}{W_1}\right)^2 + \left(\frac{\text{Error in } W_2}{W_2}\right)^2} \quad (1)$$

Since

$$\ln \left(\frac{W_1}{W_2}\right) = \frac{\Delta t \ln 2}{T_{1/2}} \quad (2)$$

Equation 1 can be converted to Equation 3.

$$\frac{\text{Error in } T_{1/2}}{T_{1/2}} = \frac{1.4427 T_{1/2}}{\Delta t} \sqrt{\left(\frac{\text{Error in } W_1}{W_1}\right)^2 + \left(\frac{\text{Error in } W_2}{W_2}\right)^2} \quad (3)$$

Equation 3 shows the error in the half-life decreases in proportion to the difference in time between two measurements and contains no exponential term as might be expected. An example of the use of Equation 3 is presented below:

Suppose the present sample is not measured again until it has decayed to such a value that accuracy of 0.01 per cent can still be obtained with the micro calorimeter. How accurate would the resulting half-life be?

The present value of 3.1260 watts is accurate to 0.01 per cent. An accuracy of 0.01 per cent can be obtained for power as low as 1000 micro-watts. Using the present half-life of 138.375 days, this would occur in 1600 days. The error in the half-life by Equation 3 would then be

$$\begin{aligned} \frac{\text{Error in } T_{1/2}}{T_{1/2}} &= \frac{1.443 \times 138.375}{1600} \sqrt{(0.0001)^2 + (0.0001)^2} \\ &= 0.0000176 \text{ or } 0.00176\% \end{aligned}$$

Thus, the error in the half-life would be 5.7 times smaller than the errors in the two power measurements. Of course, one would want numerous measurements over the entire span of time in order to show there were no other radioactive elements present.

URANIUM-234 SEPARATION

A program to separate and purify milligram quantities of uranium-234 from aged plutonium-238 solutions is in progress. The uranium is separated from the bulk of the plutonium in nitrate solution by an anion exchange process. Extraction of the uranium into (methyl iso-butyl ketone) further reduces the concentration of plutonium and most other impurities. Finally, a thenoyltrifluoroacetone (TTA) extraction removes the last traces of plutonium from the uranium. One batch of feed solution (B63) has been processed according to this scheme and 14 milligrams of greater than 95 per cent isotopically pure uranium-234 were recovered. A second batch (5H) containing 250 milligrams of uranium-234 in 26.2 grams of plutonium was processed. The product contained 70 milligrams of uranium-234 and 0.06 milligrams of plutonium.

Solution 5HC (the aqueous strip from the hexone extraction of solution 5H) was evaporated to a small volume, then diluted to approximately one normal with nitric acid. During the dilution, a large flocculent precipitate formed which could not be dissolved. The precipitate was separated by filtering and was designated 5HC-2; the supernate was designated 5HC-1. Analysis of the 5HC-1 solution indicated that it contains 7.86 milligrams of plutonium in 1.70 normal nitric acid. For the TTA extraction, the solution was diluted to 0.94

normal and hydroxylamine nitrate was added to reduce any hexavalent plutonium formed during the evaporation. The solution was extracted with half its volume of 0.46 molar TTA in xylene for ten minutes for four extractions. Extraction of the plutonium from the aqueous phase was followed by mounting and alpha counting ten-microliter samples of the aqueous phase. The extractions reduced the total alpha count by a factor of 20. Alpha pulse height analysis indicated about five per cent of the count was due to uranium-234. Additional alpha pulse height analyses were made after the fifth and seventh TTA extractions. The last analysis showed about equal counts from the uranium and plutonium. The solution was calculated to contain 70 milligrams of uranium-234 and 0.06 milligrams of plutonium-238. Further purification, especially from chemical impurities, is planned for this solution.

Previous calculations from the known age and amount of plutonium-238 in the 5H solution indicated 250 milligrams of uranium-234; this was confirmed by mass spectrometer analyses. Therefore about 180 milligrams of uranium remained to be recovered. The precipitate fraction, 5HC-2, was dissolved in concentrated nitric acid and diluted to one normal; the valence of the plutonium was adjusted, and the solution was treated with TTA-xylene in the same manner as 5HC-1. After eight extractions, when the total alpha count was reduced by a factor of 40, an alpha pulse height analysis showed no detectable uranium-234 peak. On this basis it was estimated that the solution could not contain more than a few milligrams of uranium-234.

The raffinate from the hexone extraction step (5HB) was reprocessed. Ferrous sulfamate was used as a reducing agent, and the scrub step was omitted.* The resulting aqueous strip solution was designated 5HF. The solution contained 9.3 milligrams of plutonium in 548 milliliters of 0.83 normal nitric acid. The 5HF solution was extracted twice with half its volume of TTA in xylene, which reduced the total alpha count by a factor of 45. Alpha pulse height analysis showed no detectable uranium-234 peak, which indicated that the solution could not contain more than a few milligrams of uranium-234.

The plutonium fraction from the anion exchange step and the hexone and TTA organic phases are being examined for residual uranium.

PROMETHIUM SEPARATION AND PURIFICATION

A technique is being developed to separate promethium using the HCl method.⁷ Both neodymium and samarium elute from cation exchange resins in a similar manner. This behavior was described previously (MLM-1177), but there was some question about the behavior of americium in the presence of milligram quantities of rare earth. To study this problem a substitute for the element americium was sought. Although no element will exactly duplicate the behavior of americium, bismuth was selected as an acceptable substitute.

The selection of bismuth was based on two criteria, both important in ion exchange work. The ionic radii of Bi(III) and americium(III) are not identical (americium = 1.07 Å, bismuth = 0.96 Å) but are similar enough so that parameters based on ionic radii should be similar. The ionic radii of neodymium is 0.995 Å and of samarium is 0.964 Å. Hence, the radius of bismuth is similar to the elements of interest.

An even more important parameter is the ability to form chloro complexes. In 12 normal hydrochloric acid such tendencies can be very discriminating. Both bismuth(III) and americium(III) form anionic chloro complexes; moreover, the stability constants of the 1:1 chloro complexes are not greatly different:

$$\log K^{Am} = 1.2, \quad \log K^B = 2.4$$

*Previously used extraction and stripping procedures were described in MLM-1176.

⁷K. Street and G. T. Seaborg, J. Am. Chem. Soc., 72, 2790 (1952).

The elution of bismuth from a cation exchange resin with 12 normal hydrochloric acid was studied. The elution was similar to the elution of zinc under similar conditions. Hence, it is possible to separate promethium from bismuth and zinc, and the separation should be effective for a host of other elements, including americium. On a tracer scale this is what is observed. For the present, this concludes the study of this method.

Separation of neodymium and samarium by means of 2-OH isobutyrate was also described previously (MLM-1177). The results of the separation were encouraging, but the method was not satisfactory for separating samarium from promethium. The separation was obtained by using 2-OH isobutyrate at pH 3.5. At pH 3.4, the separation is very distinct, and the difference of 0.1 pH unit is striking. Complete separation is obtained in this manner, but it is still not considered satisfactory for samarium-promethium separations. The pH sensitivity is not surprising, since rare earth 2-OH isobutyrate systems may involve species with as many as four ligands per rare earth ion. Recent literature⁹ provides an estimate of the equilibrium constants involved:

Nd (2-OH) _n	K ₁ -	420
	K ₂ -	111
	K ₃ -	25
Sm (α-OH) _n	K ₁ -	560
	K ₂ -	141
	K ₃ -	38

(This source disagrees with other sources about lactate-rare earth equilibria, casting some question on these constants). The pH sensitivity is that predicted by tracer studies of rare earths and 2-OH isobutyrate and is not unexpected.

The α-OH isobutyric acid is adjusted to the appropriate pH with ammonium hydroxide. Recent studies have indicated that the presence of an inert salt, such as NH₄Cl, in the elutant interferes with separation.

Slides of the promethium solution were mounted for counting. A 10 lambda (0.001 ml) sample of the promethium solution was diluted to 500 mls with one normal hydrochloric acid. Ten lambda (0.001 ml) was diluted to 10 milliliters and 0.5 and one milliliter aliquots were mounted. The one milliliter mount showed no alpha activity, but the 0.5 milliliter mount showed four counts per minute in a 2π geometry. The two slides showed 3.2 x 10⁴ beta and 2.6 x 10⁴ beta counts per minute respectively.

A number of reasons may account for the observed activity ratio of 1.2 instead of the predicted 2.0. The one milliliter mount spattered slightly as it was heated to vaporize the liquid. A gold slide (others were silver) bearing a count of 1.44 x 10⁵ beta counts per minute gave 23 counts per minute in good agreement with the slide bearing the 0.5 milliliter mount. Hence, for every 1.44 x 10⁵ beta counts, there are 23 alpha counts.

Alpha pulse height analysis revealed a peak at 5.47 Mev, which corresponds closely to predicted activity from americium-241. There is also a 5.32 Mev alpha whose origin is uncertain but may be curium-245 or curium-246. Gamma spectrum of the sample showed radiation at 60 kev, corresponding to americium-241. A radiation of 105 kev can also be attributed to americium-241. There are also discrete radiations at 25 kev and 45 kev. Radiation of 1.3 and 1.1 Mev may be europium-154. Other discrete radiations occur at 365 kev, 605 Kev, 865 kev, the latter two being very small. The radiations at 25 Kev and 45 kev are silver and promethium x-rays.

⁹Choppin, G. R. and Chopoorian, J. A., *Complexes of Lanthanide Elements With a Hydroxy Carboxylate With Ligands*, J. Inorg. & Nucl. Chem. 22, 97 (1961).