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J. F. Eichelberger
G. R. Grove
L. V. Jones

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These reports are not intended to constitute publication in any sense of the word. Final results either will be submitted for publication in regular professional journals or will be published in the form of MLM topical reports.

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SUMMARY

ISOTOPE SEPARATION AND PURIFICATION

Helium Superfluid Experiments A method of separating helium-3 from helium-4 in the liquid phase by a combination "superleak" and "heat flush" technique is being studied. Helium-3 enrichments to about 11% were obtained. Although the experiments are not entirely reproducible, a separation of 11% by this method is considerably higher than any results previously reported.

Carbon-13 On June 25, 1964, the first shipment of carbon-13, 13.78 g, was made to Oak Ridge National Laboratory. The carbon-13 was in the form of methane. Experiments with a single column have led to revised values of transport coefficients for the 11-column cascade. Agreement between measured and predicted values is good.

Neon The neon-22 draw-off run was completed in the processing of intermediate material used for enriching neon-21. Batch operation was started, and the column profile showed that the neon-21 had reached a concentration of about 48%.

Krypton In the initial test run to enrich the heavy krypton isotopes, krypton-84 and -86 were enriched to 92% at the bottom of the cascaded thermal diffusion column system.

Xenon

Thermal Diffusion Heavy xenon product is being drawn off at a rate of 21 ml per day from the five-stage cascaded swing system. The concentration of xenon-131 remains below 0.7%.

Gas Chromatography A separation of krypton-85 and xenon was made using Formula 676 (Pentone Co.) and dry ice as the coolant for the chromatograph column. Although this mixture was reported to give a lower temperature than that of trichloroethylene and dry ice, the separation factors were similar. Further runs will be made to determine whether the new solvent has any cooling advantage over trichloroethylene in this application.

Thermal Diffusion Research

Collision Integrals A computer program is being written to evaluate the omega integrals. The omega integrals are important not only in the evaluation of transport properties of a gas but also as a link between the theoretical molecular potential and the physical properties of a gas.

RADIOELEMENTS RESEARCH

Polonium

Decay Schemes of Polonium-208 and Polonium-209 Improvements were made in the real-to-accidental coincidence performance of the surface barrier detection system. A 3 x 3-inch sodium iodide crystal was used to replace the original 2 x 2-inch detector. A new variable geometry vacuum chamber adaptable to any of the solid state detectors in use is presently being fabricated.

The ratio of alpha activities, $\alpha\text{Po}^{208}/\alpha\text{Po}^{209}$, in a source containing both isotopes is being measured as a function of time. Alpha spectrometry results and calorimetry data yielded widely disparate results for the half-life of polonium-209.

Half-Life of Polonium-210 A calorimetric determination is being made on polonium-210 to extend the precision of the original value, which is 138.3749 days (internal probable error 0.0006 day). The weighted grand means is 138.3763 ± 0.0017 days.

The half-life of polonium-210 is being redetermined by the method of decaying pairs, primarily to test the method. The results are being compared with the highly precise calorimetric data. After 33 days and seven observations, the half-life of polonium-210 was found to be 138.7 ± 0.4 days.

Macrochemistry of Polonium The polonium electrode potentials in HCl solutions were investigated. Widely divergent results in the literature arose from the investigators' assumptions concerning the ionization of polonium in solution. The standard electrode potential was found to be -0.70 volt, calculated for the following reaction:

Actinium-227 Separation The two remaining actinium slugs of a group of three were opened and dissolved in HCl. Four elution runs were made, and the products will be combined with that of the earlier run for a final purification. The eluted fractions will be analyzed for actinium by alpha counting.

Half-Life of Thorium-228 The long-lived radioactive impurity in the thorium-228 half-life sample was identified positively as actinium-227. This identification permits a reduction of the uncertainty in the earlier half-life value. The half-life was recalculated; it is 1.91319 ± 0.00050 years.

Residue Adsorption Good separation of barium-140 and lanthanum-140 is obtained when the dried residue is converted to a fluoride and leached with water. In this case, the barium-140 is desorbed, and the lanthanum-140 is retained by a platinum surface. However, when a chloride residue on glass is leached with a mixture of ethyl or n-butyl alcohol and concentrated HCl (9:1 v/v), the lanthanum-140 is preferentially desorbed, while the barium-140 is retained by the glass surface.

ADHESIVES RESEARCH

Conductive Adhesives A previously reported conductive silicone adhesive laminate system which has a peel strength of 5.0 pounds per inch exhibited an electrical resistivity of 0.2 ohm-cm, an ultimate elongation at -65°F of 40%, and a plug tensile strength of 122 psi. This is the best silicone adhesive system evaluated to date. Substitution of silver flake for the J-2 silver used in the primer coat of this system reduced the peel strength from 5.0 to 1.5 pounds per inch.

ANALYTICAL

Polonium Urinalysis The standard method of polonium urinalysis was modified to obtain greater sensitivity of measurement. By the original method the Maximum Permissible Concentration (MPC) in the human body gave 0.95 alpha count per minute. By the new method the MPC yields 3.80 counts per minute.

ISOTOPE SEPARATION AND PURIFICATION

Processes are being developed for separating and purifying the isotopes of a number of elements including hydrogen, the noble gases, carbon, and uranium. Potential sources of supply of these materials are being evaluated.

HELIUM SUPERFLUID EXPERIMENTS

Helium-4 exhibits superfluid properties when the temperature is lowered below 2.17°K. In contrast, no evidence of superfluidity has been observed in helium-3, and probably will not be, since the nucleus of the light isotope possesses odd particle statistics as opposed to the helium-4 even particle statistics. This difference in quantum statistics accounts for the peculiar characteristics which allow the two isotopes to be separated cryogenically.

The results of previous helium isotope separation experiments could only be correlated by plotting the feed concentration of each successive experiment.¹ This curve increased uniformly from run to run, but enrichment factors and product concentrations varied without order. Therefore, two identical experiments were performed to test the reproducibility of the technique (Table 1). Two tanks of feed mixture were prepared and allowed to interdiffuse for several days. The superfluid flow was initiated with a light as in previous experiments; then the liquid level on the downstream side of the superleak was raised by applying successively increasing amounts of heat. The power input and the time applied were identical in both cases. Figure 1 shows the rate of rise of the superfluid helium-4 flowing through the leak as a function of time. The initial rise of liquid to the first level shown in Figure 1 is very rapid. The data points show the time-dependent behavior after the initial rise. The liquid level is measured by an arbitrary linear scale attached to the device inside the cryostat and is observed visually. Each scale division amounts to slightly more than 0.1 cc of liquid. The scale may be read only to the nearest division due to vibration in the liquid and to the necessarily little light inside the cryostat. Based upon observation of the liquid level of the helium-4 raffinate, the experiments were entirely reproducible within the limits of the observational error. However, one product contained 11.44% helium-3 and the other 7.93% as measured by the mass spectrometer. This variation is explainable by the fact that a difference of only one unit (about 2½% of the total raffinate) in the final level of the helium-4 raffinate can completely account for the observed difference (~4.5%) in the helium-3 product concentrations. Any slight lack of reproducibility in the quantity of helium-4 drawn through the leak results in a highly magnified discrepancy in the observed helium-3 product concentrations. The magnification factor is the overall enrichment factor of the process, in this case between 20 and 30. Thus, an analysis of results must include consideration of the reading.

Table 1
Results of Mass Spectrometer Analysis for Superfluid Separation Products

Experiment	Power Input (mw)	Ave. Time of Power Input (min.)	He ³ Content (% ± 0.02%)
I	94.4	20	Feed = 0.40
	109.7	13	Product = 11.44
	129.2	23	Dump = 0.00
	154.2	51	
	180.1	25	
II	94.4	20	Feed = 0.43
	109.7	13	Product = 7.93%
	129.2	23	Dump = 0.00
	154.2	51	
	180.1	25	

¹See previous reports in this series.

Figure 1. Rate of Rise of Helium-4 on the Downstream Side of Superleak: Comparison of Two Experiments.

In spite of the apparently inconsistent results, it is important that the helium-3 product concentrations obtained were higher than any other previously reported. Other workers have reported concentrations up to 6%. Probably, the depression of the lambda transition temperature will be the limiting factor for separation by this method. Experiments are planned to test this hypothesis.

CARBON-13

On June 25, 1964, the first shipment of carbon-13, 13.78 g, was made to Oak Ridge National Laboratory. The carbon-13 was in the form of methane ($C^{13}H_4$) which had been separated from partially enriched methane in the thermal diffusion column cascade. Details concerning the material shipped are given in Table 2. Approximately 6 g carbon-13 are still on hand.

Carbon-13 is being enriched in the 11-column, 9-stage thermal diffusion cascade at the rate of 0.025 g per day.

Experiments with a single column have led to revised values of transport coefficients for the 11-column cascade. Calculations of cascade performance were made for natural abundance feed with the new coefficients. In Table 3, the results of these calculations are compared with experimental data taken earlier. Agreement between actual and predicted cascade performance is good.

NEON

The neon-22 draw-off was stopped after 19.3 liters of neon-22 enriched to 99.94% were removed from the 48-foot thermal diffusion cascade for the enrichment of neon-21. The gas in the upper column of the cascade and gas in the reservoir and compressor was transferred into a feed tank for batch operation. This gas contained 25% neon-20, 19% neon-21, and 56% neon-22.

Table 2

**Analyses of Methane Purified at Mound Laboratory for
Shipment to Oak Ridge National Laboratory**

	Cylinder Number							
	12A432	12A451	9C8	9C9	9C10	9C11	9C12	9C13
Pressure (psia)	64.0	62.0	61.2	55.0	60.5	58.5	63.5	60.5
Carbon-13 Content (g)	4.45	4.34	0.84	0.75	0.84	0.85	0.88	0.83
Mass-17 Enrichment (%)	92.81	94.08	91.85	90.96	92.58	91.83	91.78	91.65
Carbon-13 as Percentage of Total Carbon (%)	90.36	91.85	91.29	(Est) 90.4	91.24	91.15	90.88	90.71
Methane Purity ^a (%)	98.45	97.36	96.99	96.78	96.91	96.49	97.32	97.16

^aThe major contaminant is nitrogen. Small quantities of carbon dioxide, oxygen, argon, and some higher hydrocarbons may also be present.

Table 3

**Measured and Calculated Composition Profiles and Production Rates
for the 11-Column, 9-Stage Methane Thermal Diffusion Cascade**

	Mass-17 Enrichment (%)		
	2-5-64	2-28-64	Calculated
Waste Stream	- -	- -	0.92
Feed	1.09	- -	1.1
Stage 1, Bottom	1.38	1.32	1.28
Stage 2, Bottom	2.85	2.56	2.11
Stage 3, Bottom	4.26	3.53	2.57
Stage 4, Bottom	7.25	5.79	4.25
Stage 5, Bottom	19.10	14.20	11.44
Stage 6, Bottom	50.46	40.52	40.62
Stage 7, Bottom	93.04	77.17	81.24
Stage 8, Bottom (Product Withdrawal)	95.98	95.94	96.00
Stage 9, Middle	97.06	96.87	- -
Waste Rate (g/day)	25.0	25.0	26.4
Product Rate (g/day)	0.03	0.05	0.0493

The batch system filled with this gas consisted of two 24-foot columns connected in cascade by a bellows pump for circulation of the gas. After 24 hours of static operation with a wire temperature of 800°C the column profile shown as Curve A in Figure 2 resulted.

Figure 2 shows that neon-22 enriched above 99.0% can be removed from the bottom of the cascade. The procedure is to remove 100 to 200 cc every other day from the system and then to refill the column to operating pressure from the feed tank. After 400 cc had been removed, the column profile changed as shown by Curve B in Figure 2, indicating a neon-21 concentration of about 48%.

KRYPTON

A two-stage thermal diffusion cascade is being used to enrich the heavy isotopes, krypton-84 and -86. The initial equilibration tests showed a concentration of about 92% krypton-84 and -86 at the bottom of the cascade at a system pressure of about 300 torr. The pressure is being increased to improve the separation.

XENON

Thermal Diffusion In a five-stage cascaded swing system, the maximum flow rate of less than 1% xenon-131 product starting with fission xenon is being experimentally determined. The initial withdrawal rate of 24 ml every two days was increased to 21 ml per day. The product concentration of xenon-131 remains at less than 0.7%.

Gas Chromatography Xenon obtained as a reactor off-gas contains radioactive krypton-85 as an impurity. This impurity is removed before the xenon isotopes are enriched. An experimental chromatograph was built to study the parameters affecting the krypton-xenon separation. Studies made with the experimental chromatograph are used in determining modifications in design and in operational techniques of the chromatograph.

It was found in past separations that decreased column temperatures yield increasing values for cleanup:

Figure 2. Curve A is the Neon-21 Profile 24 Hours After Feed Containing 18.7% Neon-21 Was Put Into the Columns; Curve B is the Profile After 400 cc Neon-21 Were Drawn off at the Bottom of the Cascade.

$$\text{Cleanup Factor} = \frac{\text{Kr}^{85} \text{ in feed}}{\text{Kr}^{85} \text{ in Xe trap}}$$

Further, increasing the column temperature at some preselected intermediary point in the separation gave higher cleanups than isothermal separations. The best separations in the past were made with 195°K and 243°K as the column temperatures. A trichloroethylene - dry ice mixture was used for operation at 195°K.

The Pentone Company claims their proprietary solvent, Formula 676, and dry ice can be used to reach 186°K. A separation made with Formula 676 and dry ice as the low temperature bath showed no detectable difference from those separations made with trichloroethylene and dry ice. The run was made with an internally finned column with a cross-sectional area of 5.5 cm² and a length of 19 cm. The adsorbent was activated coconut charcoal, 50/60 mesh. The column temperature was increased to 243°K when the krypton-85 peak reached its maximum value; the xenon trap replaced the krypton trap after 40 minutes of running time. The krypton-85 retention time was 13 minutes as in previous runs in which the column temperatures were 195°K and 243°K. The cleanup was 10,600, the same order of magnitude as in separations made at 195°K and 243°K. Although this run indicated that Formula 676 does not offer any advantage over trichloroethylene, more runs will be made for verification.

THERMAL DIFFUSION RESEARCH

Collision Integrals The omega integrals arise from the Chapman-Enskog solutions to the Boltzman equation. These integrals, defined in terms of the intermolecular potential, are used in the expressions for the transport properties of a gas. Thus, the omega integrals are important not only in the evaluation of the transport properties, but also as a link between the theoretical molecular potential and the physical properties of a gas.

To evaluate these integrals, an intermolecular potential of the form

$$Ae^{-ar} - Ce^{-cr} - B_1/r^6 - B_2/r^8 - B_3/r^{10}$$

was assumed, where the constants A , a , C , c , B_1 , B_2 , B_3 are determined from quantum mechanical calculations and experimental data. Computer programs using this form of the potential are being written to numerically evaluate the omega integral for helium and argon. The helium potential will be obtained by analyzing all the available potentials. The argon potential has been chosen as that of Mason² so that the results can be compared with those in Hirschfelder.³

²E. A. Mason, Jr., *J. Chem. Phys.*, 22 (1954).

³J. O. Hirschfelder, C. F. Curtiss, and R. B. Bird, *Molecular Theory of Gases and Liquids*, New York, John Wiley and Sons, 1954.

RADIOELEMENTS RESEARCH

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

POLONIUM

Decay Schemes of Polonium-208 and Polonium-209 Additional improvements have been made in the real-to-accidental coincidence performance of the surface barrier detection system through (1) installation of a 3 x 3-inch NaI detector in place of the 2 x 2-inch detector originally employed, (2) more efficient shielding of gamma rays, and (3) more precise time balance of the alpha and gamma channels to reduce the resolving time into the 30-40 nanosecond range.

A summary of earlier alpha-gamma coincidence studies using a 100 mm² ORTEC barrier detector and 2 x 2-inch detector are shown in Table 4. Although the energy calibrator of the system was not entirely satisfactory, it appears that there are two low energy, low abundance groups present. These groups have energies of about 4.21 and 4.25 Mev. The 4.25-Mev group appears to be in coincidence with gamma rays in the 200 to 400-kev range, whereas the lower energy group appears to be in coincidence with one or more gamma rays in the 800 to 950-kev range. As described in a previous report these groups are tentatively associated with polonium-209 and polonium-208 decay, respectively. These results give rise to the tentative decay schemes shown in a previous report.⁴ Although the results are not shown in Table 4, the 4.62-Mev alpha group is prominent in all spectra using gamma-ray coincidence gates below 260 kev.

Table 4
Summary of Coincidence and Energy Measurements
Using 100 mm² ORTEC Barrier Detector

Gamma-Ray Gate Condition Imposed (kev)	Length of Measurement (min)	Energy of Low Abundance Group (Mev)	Net Coincidence per 1000 Min
120-200	1000	4.25	77
200-350	1000	4.27	321
120-400	5000	4.26	310
400-670	5000	4.24	11
800-950	5000	4.20	21

A new, variable geometry vacuum chamber adaptable to any of the solid state detectors in use is presently being fabricated. A cross-section view of the chamber is shown in Figure 3. This chamber will permit any of the solid state detectors to be brought down next to the source, yielding high geometry for both alpha and gamma detectors.

Alpha pulse-height analyses are being made on a source containing polonium-208 and polonium-209. The ratio of the alpha activities, $\alpha\text{Po}^{208}/\alpha\text{Po}^{209}$, is being measured as a function of time.⁵ All the counting data

⁴J. F. Eichelberger, G. R. Grove, and L. V. Jones, *Mound Laboratory Progress Report for April, 1964*, MLM-1196 (July 10, 1964), pp. 7-10.

⁵J. F. Eichelberger, G. R. Grove, and L. V. Jones, *Mound Laboratory Progress Report for February, 1964*, MLM-1185 (April 14, 1964), pp. 9-10.

Figure 3. Aluminum, Variable Geometry, Solid State Vacuum Chamber.

have been carefully reevaluated to take into account the contribution of low-energy tailing of the polonium-208 alpha group into the polonium-209 alpha group. The decay constant of the ratio was evaluated by a weighted least-squares method, and the half-life of polonium-209 was found to be 37.4 ± 7.7 years, in good agreement with previously reported values.

The $\text{Po}^{208}/\text{Po}^{209}$ source whose decay has been observed for about 12 years by calorimetry was opened, sampled, and resealed in a tantalum capsule. The ratio of the alpha activities, $a\text{Po}^{208}/a\text{Po}^{209}$, was determined by alpha pulse-height analysis to be 11.577 ± 0.024 on June 18, 1964. This value, when combined with the calorimetry data, gives a half-life of polonium-209 in excess of 200 years. Further critical analyses of the alpha spectrometry results and the calorimetry data are required to reconcile the wide disagreement in the various values of the polonium-209 half-life.

Half-life of Polonium-210

Calorimetry A determination of the half-life of polonium-210 is being made with a sample fabricated in November, 1963.⁶ The goal is an order of magnitude more precision than the previous determination of 138.3749 days which had an internal probable error of 0.0006 day, or one minute. The sample has been assayed 78 times in Calorimeter 90 since November 6, 1963, when it produced 3.127 watts of power. On March 23, 1964, measurements were initiated in Calorimeter 91. This sample will be assayed periodically in both calorimeters until June, 1966, then measurements will be continued in Micro-calorimeters 60 and 67 for another year or more.

⁶J. F. Eichelberger, G. R. Grove and L. V. Jones, *Mound Laboratory Progress Report for November, 1963*, MLM-1178 (November 30, 1963), pp. 22-23.

The complete half-life results to date are given in Table 5.

Table 5
Half-life of Polonium-210

Sample	Calorimeter	Time Span	Observations	Initial Power (watts)	Half-life (days)	Internal Probable Error (days)
1	39 & 60	Aug. 1954 to May 1958	47	2.6556	138.3749	0.0006
2	90	Nov. 1963 to June 1964	78	3.127	138.3832	0.0011
3	91	March 1964 to July 1964	19	1.566	138.3726	0.0015
Weighted Grand Means					138.3763±0.0017	

Also included are the results of the previous determination. The three values were combined by giving each a weight inversely proportional to the square of its internal probable error. The result is 138.3763 days with a probable error of 0.0017 day as calculated from the external consistency of the three values. The means are not consistent by the test given by Birge, but the variation is probable due to the processing of the data with a high degree of precision. That the results with Calorimeter 90 are not as good as expected can be attributed to the establishing of techniques during the course of the measurements, and to the room humidity which was not controlled during the entire experiment. On the other hand the results with Calorimeter 91 are better than were expected. The sample has not decayed to the most precise range of the calorimeters, which is between 0.6 and 0.3 watt. In this range the probable error per observation should be 0.001%, which is equivalent to three minutes of decay by polonium-210. The short time stability of both calorimeters is such that one minute produces a detectable decrease in bridge potential as indicated on the recorder.

Half-life Determination by Decaying Pairs The half-life of polonium-210 is being redetermined by the method of decaying pairs⁷ primarily to test the method and to examine certain parameters. Polonium-210 was selected because its half-life is known with high precision and accuracy from calorimetric data.⁸

Polonium-210 samples were prepared by electrodepositing polonium on platinum foil and covering the deposits with vacuum evaporated gold to a thickness of approximately one tenth of an alpha range.

Six samples, whose initial alpha counting rates ranged from 7×10^4 to approximately 1×10^6 counts per minute are being counted periodically, and the data are being analyzed by the IBM 1620 computer. After 33 days and seven observations, the half-life of polonium-210 was found to be 138.7 ± 0.4 days, as compared with the accepted literature value of 138.401 ± 0.006 days.

⁷J. F. Eichelberger, *Mound Laboratory Monthly Progress Report for December, 1961*, MLM-1128, (December 29, 1961), p. 11.

⁸J. F. Eichelberger, K. C. Jordan, S. R. Orr, and J. R. Parks, *Phys. Rev.*, **96**, 719 (1954); see also *Calorimetry* above.

Macrochemistry of Polonium Several experiments were performed in an investigation of the polonium electrode potentials in HCl solution. Bagnall⁹ and Payne and Nelson,¹⁰ using polonium-210, gave the following results for the several polonium electrode reactions in HCl:

Electrode	E°, Ref. 9 (v)	E°, Ref. 10 (v)
Po, Po(IV) (HCl)	-0.55	-0.654
Po, Po(II) (HCl)	-0.38 (calculated)	-0.560
Po(II), Po(IV) (HCl)	-0.72	-0.748

The large discrepancies in the Po, Po(IV) and Po, Po(II) results arise from the assumptions made by the investigators: Bagnall assumed that polonium dissolved in HCl to give Po(IV); whereas, Payne and Nelson supposed that the dissolution first gave Po(II) and then Po(IV). If Bagnall's results are recomputed, assuming the first reaction to be $\text{Po} = \text{Po(II)}$, the potentials agree very well with those of Payne and Nelson. In either case, the potentials of the polonium electrodes are consistent with chemical behavior of polonium in HCl.

The experiments reported above were performed with polonium-210, and the results may have been affected by the severe alpha radiolysis of the solution caused by the decay of polonium-210.

The use of the mixed isotopes polonium-208 and polonium-209 lessens the effects of alpha radiolysis because of the longer half-lives of these isotopes. In the experiments reported here the potential of a platinum or gold electrode immersed in a HCl solution of $\text{Po}^{208}/\text{Po}^{209}$ was measured against a saturated calomel electrode electrically connected to the polonium solution by means of a potassium chloride-agar salt bridge. The yellow Po(IV) solution was treated with two reducing agents: stannous chloride, to produce polonium metal; or hydrazine, which produced Po(II). Due to the oxidizing effect of alpha radiolysis in the systems, the excess reducing agent was gradually oxidized; then the reduced polonium was slowly oxidized. The cell potential was recorded as a function of time. Inflections in the potential vs. time curve were obtained and indicated that oxidation of the polonium from lower to higher oxidation states was taking place. These inflections could be correlated with color changes in the solution. Polonium metal was seen as a cloudy grey suspension; Po(II) was observed to be light pink; and Po(IV) had the characteristic yellow color of oxidized polonium in hydrochloric acid.

The results of several experiments showed that the electrode potential for the oxidation of polonium metal to Po(II) and HCl was generally obscured by other reactions, probably due to noble metal impurities. The electrode potential of the $\text{Po(II)} \rightarrow \text{Po(IV)}$ oxidation after reduction with hydrazine was usually clear cut, however. The standard electrode potential was found to be -0.70 volt, calculated for the reaction

Further work is in progress to determine the chloride-ion dependence of this reaction. The polonium is being repurified for additional experiments on the oxidation of polonium metal.

ACTINIUM-227 SEPARATION

Both actinium slugs remaining to be processed¹¹ contained some radium-226. These slugs were opened, and the contents were dissolved in 2 M HCl. The solution was processed through the ion exchange column in

⁹K. W. Bagnall and J. H. Freeman, *J. Chem. Soc.*, 2770 (1956).

¹⁰H. V. Moyer (ed.), *Polonium*, USAEC Report TID-5221 (1956), p. 80.

¹¹J. F. Eichelberger, G. R. Grove and L. V. Jones, *Mound Laboratory Progress Report for June, 1964*, MLM-1204, (June 22, 1964), p. 19.

four runs. Runs 2 and 3 were similar to Run 1, with the same problem of gas formation in the resin column. During Run 4 the water jacket heating element burned out. Due to the lower operating temperature, the equilibrium between the metal ions absorbed on the resin and those in solution shifted unfavorably, resulting in greater overlap of the elution bands. A new column with larger resin particles (20-40 mesh compared to 100-200 mesh) of the Dowex resin AG50W-X8 was placed in the hot cell for Run 5. The larger resin particles were used to increase the flow rate and to reduce the problem of gas formation in the resin. However, the greater flow rate and decreased surface area of the resin resulted in some solution passing through the resin without complete absorption of the actinium. In the other four runs actinium started to elute prematurely in the 2 M HNO₃ fractions. This is probably the result of the larger than normal volumes of eluent used to insure removal of the impurities before the actinium.

All the fractions containing actinium (as determined by alpha pulse height analysis) are being combined for a final purification run.

It was not possible to distinguish between the radioactive isotopes being eluted by picking up the characteristic gamma energies in the collimator detector crystal and analyzing the energy spectrum with a single-channel analyzer. Part of the cause may have been the reduction in energy of some of the gamma from scattering.

Since the fractions could not be analyzed by gamma counting through the collimator tube, it was necessary to sample each fraction, remove the sample from the hot cell, and prepare each for alpha energy analysis. Since the alpha energy of actinium-227 is considerably lower than that of any of the daughter products, this isotope can easily be distinguished from its daughters. Radioactive impurities in the actinium-227 also have more than 10⁴ times as many alpha particles emitted per gram molecular weight per unit time compared to actinium and, therefore, are easily detected in the presence of much larger quantities of the actinium-227 isotope. Final purity and total actinium purified will be determined by calorimetry.

HALF-LIFE OF THORIUM-228

The half-life sample of thorium-228, which has been assayed periodically since 1954, was opened in the new cave facility; the remaining activity was examined with an alpha pulse height analyzer. The long-lived radioactive impurity was identified positively as actinium-227. This knowledge can be used to reduce the uncertainty in the half-life results presented in an earlier report.¹² Table 2 of that report lists the half-life of thorium-228 as 1.91319 years, or 1.91443 years, depending on whether the impurity was actinium-227 or radium-225. The half-life can now be reported as 1.91319 ± 0.00050 years, where the limits are internal probable errors. An indication of the high reliability of this result is obtained by comparing the alpha ratio of thorium-228 to actinium-227 as calculated from Table 2 of the referenced report to the ratio obtained with the pulse height analyzer. The ratios as of June 29, 1964, are 25.9 and 25.6, respectively. The watts per curie values used to obtain the alpha ratio from the calorimeter data were 0.2016 for thorium-228 and 0.2013 for actinium-227.

RESIDUE ADSORPTION

Residue adsorption studies with barium-140/lanthanum-140 have been resumed. A new batch of barium-140 received from Oak Ridge National Laboratory has been purified of calcium, the principal impurity, by cation exchange with a Dowex-50 resin column.

The solution of barium-140 and lanthanum-140 was loaded on the column in 1 N HCl, and the column was eluted with 3 N HCl. The effluent was collected in small fractions, of which the ninth through twelfth fractions

¹²J. F. Eichelberger, G. R. Grove, and L. V. Jones, *Mound Laboratory Progress Report for July, 1963*, MLM-1170 (July 30, 1963), pp. 13-15.

contained approximately 95% of the barium 140. Atomic absorption analysis showed that the calcium impurity was concentrated in the fifth and sixth fractions, indicating a good separation.

The four fractions containing the bulk of the barium 140 were combined and diluted with deionized water to 1 N HCl. The new stock solution is being stored in a polyethylene bottle to eliminate the possibility of re-introducing inorganic impurities from radiolytic attack on borosilicate glassware.

In the residue adsorption work with strontium/yttrium 90, it was found that hydroxide residues leached with water left approximately 90% of the yttrium and 10 to 15% of the strontium adsorbed. The separations of barium and lanthanum-140 hydroxides have not been as satisfactory.

Although lanthanum-140 hydroxide appears to behave very much like yttrium 90 hydroxide, in that approximately 95% of the lanthanum remains adsorbed when the dried residue is washed with water between 35 and 45% of the barium 140 is also retained by a platinum surface. Several possible explanations are being studied, including the difference in the specific activities of strontium-90 and barium 140, the possibility that trace sulfate from the ion exchange resin is causing the barium-140 to be adsorbed as a sulfate, and the difference in the relative solubilities of strontium and barium carbonates.

A satisfactory separation is obtained when the barium/lanthanum-140 is dried as a fluoride and leached with water. The result of one such separation is shown in Figure 4, where the platinum surface retained only 0.9% of the barium-140 while retaining 89.3% of the lanthanum-140. The upper spectrum is that of the residual lanthanum-140 showing the absence of the 162-kev photopeak of barium-140. The amount of lanthanum-140 desorbed with the barium-140 is indicated in the lower spectrum by the relative intensities of the photopeaks at 0.83 and 1.6 Mev.

The order of desorption is reversed if the residue is converted to a sulfate and leached with a dilute solution of H_2SO_4 . In one such separation, the barium/lanthanum-140 was dried in a glass vial with 0.25 ml of 0.002 N H_2SO_4 and leached with 1 N H_2SO_4 . The glass retained 48.0% of the lanthanum-140 and 64.2% of the barium-140.

A much better separation with reverse order desorption is obtained if the dried residue is leached with non-aqueous solvents:

One drop (0.05 ml) of the 1 N HCl solution of barium/lanthanum-140 was evaporated to dryness in a glass vial at 95°C. The vial was cooled to 25°C and leached twice with a mixture of n-butyl alcohol and concentrated HCl (9:1 v/v). The vial retained 9.4% of the lanthanum-140 and 72.4% of the barium-140. Similar results were obtained with ethyl alcohol as the non-aqueous component and with concentrated HNO_3 as the aqueous component.

The significance of this reversal of desorption order is not yet clear; there is a possible relationship between this phenomenon and the classical separation method in which barium chloride is precipitated from a 6:1 mixture of concentrated HCl and ether. In this case, lanthanum-140 remains quantitatively in solution.

The adsorption behavior of barium/lanthanum-140 in other nonaqueous media is being investigated.

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Figure 4. Separation of Barium-140 and Lanthanum-140 From a Fluoride Residue.

ADHESIVE RESEARCH

Adhesive formulations are being developed for specialized applications. The adhesives must have satisfactory working and curing characteristics, they must remain flexible at low temperatures and retain adequate tensile strength at elevated temperatures.

CONDUCTIVE ADHESIVES

The previously reported¹³ conductive adhesive laminate system which has a peel strength of 5.0 pounds per inch exhibited an electrical resistivity of 0.2 ohm-cm one month after cure, an ultimate elongation at -65°F of 40%, and a plug tensile strength at room temperature of 122 psi. This system is the best silicone adhesive formulation evaluated to date. Substitution of silver flake for the J-2 silver used in the primer coat of this system reduced the peel strength from 5.0 to 1.5 pounds per inch.

¹³Eichelberger, MLM-1204, p. 22

ANALYTICAL

Methods of analyzing elements and compounds are being developed to support other programs at Mound Laboratory. These methods include instrumental techniques and classical wet methods.

POLONIUM URINALYSIS

For the past 16 years the polonium urinalysis process has been essentially that reported by E. S. Spoerl.¹⁴ This procedure consisted of placing 50 ml of urine in a paper cup to which HCl was added. The polonium was plated onto a one-inch copper disk suspended in the solution which was stirred by a mechanical stirrer for a period of two hours.

On July 13, 1964, a new procedure was adopted which employs a Lucite deposition cell. This cell uses a 1-7/8-inch stainless steel disk which exposes only one surface to the acidified urine solution for spontaneous deposition of the polonium. The volume of urine for analysis was increased from 50 ml to 100 ml.

The analytical procedure is essentially the same as before, but the mechanics of the new method substantially improved the sensitivity. This increase in sensitivity was gained by

- (a) Plating one surface rather than two as in the old method, and
- (b) Using twice the volume of urine.

The combined effect is that for a person having the Maximum Permissible Concentration (MPC) based on the 0.04 microcurie per total body; thus, the rate would be 0.95 count per minute. With the new method the MPC yields 3.80 counts per minute. This greater amount allows a much shorter counting time than was possible with the old method. This new method also makes possible the detection of fractional body burdens which would have been extremely difficult to detect with the old method.

¹⁴E. S. Spoerl, *Urine Analysis Procedures at Mound Laboratory*, MLM-460 (April 14, 1950).