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SUMMARY

Radioelements

Uranium-234 Separation and Purification Mound Laboratory is separating and purifying uranium-234 from aged plutonium-238 solutions by a combination of ion exchange and solvent extraction techniques. Two aged plutonium samples have been processed and 84 milligrams of uranium-234 were recovered. A third source containing two grams of uranium-234 is being processed. In the first two anion exchange runs, the uranium recovery was 40 to 50%. A method for the determination of uranium in the presence of plutonium, iron, and aluminum was developed.

Polonium-210 in Nature The polonium-210 content of cigarettes and pipe tobacco was determined to be 0.30 picocurie (10^{-12} curie) per gram, in good agreement with recently reported values. The polonium-210 probably results from decay of naturally occurring uranium-238 and radium-226.

Polonium-209 Half-life The half-life of polonium-209 was determined by determining the alpha activity ratio of polonium-208 to polonium-209 as a function of time in a sample containing both isotopes. The half-life was found to be 42.4 ± 6.0 years, in fair agreement with the previously reported value of 40.2 ± 2.6 years.

The half-life of the metastable state at 0.305 Mev in arsenic-75 was determined by a coincidence method to be 15.64 ± 0.92 milliseconds. The same coincidence technique was used in an unsuccessful attempt to detect a metastable state at 1.42 Mev in bismuth-208, which may be populated in the electron-capture decay of polonium-208.

Decay Scheme Studies From a study of the alpha-gamma coincidence spectrum of polonium-208 and polonium-209, two additional alpha groups have been tentatively established at 4.62 and 4.78 Mev.

Residue Adsorption Difficulty is still being experienced in preparing a barium-140 stock solution free of calcium.

Half-life of Radium-223 The results of treating alpha-counting data by two different computational methods are compared with the calorimetric results. The computed values are found to be in good agreement.

Viscosity of Plutonium Alloy The viscosity of a molten plutonium-one weight % gallium alloy was measured at 12 temperatures from its 669° liquidus temperature to 816°. The viscosity decreased from 8.82 centipoise at the liquidus temperature to 6.60 centipoise. The one weight % addition of gallium to plutonium raised the viscosity an average of 50% as compared to unalloyed molten plutonium in the same temperature interval.

Alpha and Neutron Sources

The increase in the neutron flux of plutonium-beryllium sources due to the ingrowth of americium-241 is being measured.

Isotope Separation

Carbon-13 Using the 11-column thermal diffusion column cascade a production rate of 0.6 gram carbon-13 per month was established.

Thermal Diffusion Theory A rigorous theory of a thermal diffusion column is presently being developed. The dependence of the coefficient of viscosity (η), thermal conductivity (λ), and density (ρ) on the composition of the isotopic mixture, which was neglected in the Jones and Furry theory, is being taken into account.

The revised theory allows for a variation in wire temperature, a phenomena which has been observed experimentally and which arises because of the concentration dependence of the thermal conductivity.

Plastics and Adhesives Research

Plastics Heat resistant, epoxy foam formulations were evaluated. One formulation was superior in solvent resistance and compressive strength, but was not as resistant to heat as one of the other formulations.

Adhesives Three manganese dioxide-cured, polysulfide-based conductive adhesives were prepared and evaluated. The electrical resistivities of the formulations ranged between 0.05 and 0.06 ohm-cm; tensile bond strengths ranged between 280 and 300 psi, and peel strengths ranged between six and seven pounds per inch. Films of the formulations had an ultimate elongation between 1- and 11% at -65°F. Substitution of polysulfide prepolymer LP-3 and LP-33 in these formulations did not appreciably change their handling properties.

The silicone potting compound, LTV-602, was reinvestigated as a base for conductive adhesives. All formulations had low peel strengths of about 0.5 pounds per inch. The electrical resistivities of these formulations increased on aging. These were the most flexible materials at -65°F tested to date

Analytical

The construction of Calorimeter 91 was completed and tested. Calorimeter 91 is an automated precise twin-bridge calorimeter designed to accurately assay radioactive materials.

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.

URANIUM-234 SEPARATION AND PURIFICATION

Milligram quantities of uranium-234 are being separated from aged plutonium-238 solutions. The uranium is separated from the bulk of the plutonium in nitrate solution by an anion exchange process. Extraction of the uranium into hexone (methyl iso-butyl ketone) further reduces the concentration of plutonium and most other impurities. Finally, thenoyltrifluoroacetone (TTA) extraction removes the last traces of plutonium from the uranium.

Two aged plutonium samples were processed using this scheme and 84 milligrams of uranium-234 were recovered. A third sample containing two grams of uranium-234 is being processed. In the first two anion exchange runs, the uranium recovery was 40 to 50%. A method for the determination of uranium in the presence of plutonium, iron and aluminum was developed, and the behavior of uranium on an anion exchange column was investigated.

The absorption of uranium on an anion exchange resin from a high nitrate salt, low acid media is being studied. In a run using Dowex 1X10 instead of Dowex 1X4, 88% of the uranium was recovered. The low recoveries were caused by the relatively slow absorption rate of the uranium in this system. In a run using Dowex 1X4 in which the flow rate was reduced to two milliliters per minute, 99% of the uranium was recovered. Uranium was analyzed by the indirect PAN method.¹

A uranium product from one of the earlier runs was analyzed for aluminum by emission spectroscopy and indicated negligible aluminum contamination. Therefore the following scheme to purify uranium was proposed:

1. Adjust the uranium containing sample to 1.6 M in $\text{Al}(\text{NO}_3)_3$ and 0.3 M in HNO_3 . Add a reducing agent (such as ferrous sulfamate) to reduce any plutonium to the non-absorbed trivalent state.
2. Slowly pass the solution through a Dowex 1X4 column to absorb the uranium. Wash with $\text{Al}(\text{NO}_3)_3\text{-HNO}_3$ solution to remove any loosely held impurities. Wash with 8 M HCl to remove the aluminum.
3. Elute the uranium with 0.1 N HCl .

The anion exchange behavior of uranium was investigated in a 2 cm diameter column, 30 cm high, containing 110 ml of Dowex 1X4 resin. The resin was preconditioned with five column volumes of 7.2 N HNO_3 . A feed solution containing 86.3 mg of uranium in 400 ml of 7.2 N HNO_3 was loaded on the resin at a flow rate of 5 ml per minute. The resin was washed with 400 ml of 7.2 N HNO_3 at 7 ml per minute. The resin was eluted with 500 ml of 0.35 N HNO_3 at 10 ml per minute. Fifty milliliter samples of the effluents were collected and analyzed for uranium by the indirect PAN method.

A total of 83.74 mg of uranium, or 97.0% was recovered. The data indicate that several column volumes of 7.2 N HNO_3 are required to wash the uranium off the column. While these data are not directly applicable to mixtures of plutonium and uranium, this system will be useful in studying techniques of accelerating the removal of uranium.

¹G. R. Grove, L. V. Jones, and J. F. Eichelberger "Mound Laboratory Progress Report for January, 1964" MLM-1183

Solutions containing 56.76 grams of plutonium were divided equally among three bottles designated S3-1A, S3-1B, and S3-1C. Absorption spectra of these solutions showed peaks at 820 millimicrons, characteristic of plutonium(VI). Sixty milliliters of two-molar hydrazine nitrate solution was added to each bottle of S3-1A and S3-1B to reduce the plutonium. The solutions were boiled gently for one hour. The blue color of plutonium(III) was not observed; however, the spectra showed that most, but not all, of the plutonium(VI) had been reduced. Solutions S3-1A and S3-1B were used as feed solutions for the first ion exchange run, and their characteristics are listed below:

	<u>S3-1A</u>	<u>S3-1B</u>
Volume (ml)	718	818
N HNO ₃	7.4	6.7
Pu (g)		
Total mg/ml	18.92	18.92
	26.35	23.12
U (mg)		
Total mg/ml	197	197
	0.274	0.241

(Plutonium determined by alpha counting, uranium determined by the modified PAN method).

The ion exchange column was 4.0 cm in diameter (12.56 cm² cross section) and about 45 cm high. The capacity of the column was slightly over 600 ml with an additional 500 ml capacity in a reservoir.

The Dowex 1X4 resin previously had gone through two 2 N NaOH-2 N HNO₃ cycles and had been stored in 2 N HNO₃. The column was loaded with about 550 ml of resin and backwashed to remove the fines. The resin was conditioned with one liter of 7.2 N HNO₃ and the feed solutions passed through at rates of 5-10 ml per minute. The feed was continued past the breakthrough point, then 700 ml 7 N HNO₃ was used to wash the column. The plutonium was eluted with about three liters of 0.35 N HNO₃. Hydroxylamine nitrate solution helped remove the last traces of plutonium. The elutriates, containing the plutonium, were evaporated to less than a liter before sampling. The various solutions resulting from Run I are listed in Table 1 with analytical results and comments.

Table 1
RESULTS OF ION EXCHANGE RUN I

Solution	Volume (ml)	Pu (gm)	U (mg)	Comments
S3-1R1	944	0.004	3	First raffinate, resin 80% loaded.
S3-1R2	780	0.661	86	Second raffinate, resin loaded.
S3-1R3	626	0.470	126	Wash solution and elute until Pu appeared.
S3-13	769*	33.13	317	Elute

*Volume after evaporation and at time of sampling.

(Plutonium determined by alpha counting, uranium determined by modified PAN method).

The behavior of the plutonium is similar to that observed in earlier work. The first raffinate solution contained little plutonium while the second raffinate and wash solutions, collected during and after the breakthrough point, contain appreciable quantities of plutonium. However, the behavior of the uranium was unexpected. Previous information indicated that the uranium could not successfully compete with the plutonium for the active sites on the resin. While the resin was not loaded to capacity with plutonium, it was felt that the uranium should be washed off with a minimum of 7.2 N HNO₃. In addition, the wash solution (S3-IR3) was collected up to the point where the plutonium began to elute with dilute HNO₃. The uranium should be eluted easier than the plutonium, and thus should be washed off before the plutonium elutes. However 60% of the uranium remained with the plutonium (S3-IE).

The possibility of uranium being in a state which is strongly held by the resin (e.g. quadrivalent oxidation state or an anionic complex) seems unlikely. The uranium may have eluted first but was mixed with the first part of the plutonium elution. The mass balance of the uranium showed a gain of 139 mg of uranium. Part of this is believed to be due to small errors in the analytical method being magnified by dilution factors. The mass balance of the plutonium showed a loss of 3.58 grams. Again part of this may be due to sampling and counting errors, but also some plutonium may have remained on the resin.

The plutonium fraction from Run I (S3-IE) contained enough uranium to make preprocessing profitable. The solution was diluted to nearly 1.9 liters and adjusted to 7.2 N in HNO₃. An absorption spectrum of this solution showed a small peak for plutonium(VI) and the reduction step was omitted. The procedure, columns, and resin were the same as used for Run I. Following the feed solution, about one liter of 7.2 N HNO₃ was used as a wash. Again the raffinates were collected until the plutonium started to elute from the bottom of the column. In addition, the first 200 ml of elutriate were collected and held separate from the remainder of the elutriates to determine how much uranium was eluting with the first part of the plutonium. About four liters of dilute HNO₃ were required for elution. These elution solutions are awaiting evaporation and analysis. The analytical results of Run II are given in Table 2.

Table 2
RESULTS OF ION EXCHANGE RUN II

Solution	Vol. (ml)	Pu (g)	U (mg)	Comments
S3-IIR1	911	0.005	nil	First raffinate, resin 60% loaded.
S3-IIR2	924	0.017	23.8	Second raffinate, resin 90% loaded.
S3-IIR3	927	0.272	123	Some breakthrough, mainly 7.2 N wash
S3-IIR4	405	0.256	47	7.2 N wash add 0.35 N until Pu appeared.
S3-IIE1	212	5.62	38	First part of elution.
S3-IIE2	(Analytical results not yet available).			

(Plutonium determined by alpha counting, uranium determined by modified PAN method).

Uranium recovery in Run II was better (50% in raffinates) than Run I (40%) but it was still not satisfactory. In Run II an additional 10% of the uranium appeared with the first part of the plutonium elutriate, but presumably the remaining 40% again followed the plutonium. Apparently, larger quantities of wash solution (perhaps several liters) are required to separate the uranium from the plutonium.

Less than one third of the uranium was recovered in the processing of the 5H material reported earlier. The plutonium fraction from this processing was analyzed by the modified PAN method and found to contain 140 mg of uranium. This, plus the 70 mg recovered, represent the major fraction of the original uranium.

POLONIUM-210 IN NATURE

An article by Radford and Hunt² reported that cigarettes contained 0.43 picocurie (10^{-12} curie) of polonium-210 per cigarette. The conclusion of these authors was that "...Po²¹⁰ inhaled in cigarette smoke may act as an important initiator in the production of bronchogenic carcinoma". The experimental work was verified, and the work was extended to the determination of polonium-210 in pipe tobacco.

The polonium-210 content of cigarettes and tobacco was determined by treating ten-gram batches (approximately ten cigarettes) with HCl and heating until a homogeneous, dark brown mixture was obtained. The tobacco was weighed out as received; that is, directly from pack or pouch, with no attempt to control or estimate the moisture content.

The HCl solution was diluted to 6 M, and the polonium was plated by spontaneous electrodeposition on Krylon-masked silver foils at 80-90°C. Chemical yields were determined by spiking the tobacco solution with a polonium-208 tracer containing 5.12 ± 0.50 alpha counts per minute. The total alpha activity of the foils was measured in low background (3 - 5 counts per hour) alpha counters. The polonium-208 yield could then be determined by alpha pulse-height analysis on the plated sample.

To check the reliability of the determination of the chemical yield, several platings were made on each batch. In every case, the polonium-208 and polonium-210 activity ratios agreed within experimental error. This indicates that there was no gradual leaching of the polonium-210 from the sample, and also showed that the samples were not contaminated from laboratory sources of polonium-208 and polonium-210.

The results obtained are summarized below:

Sample	Po ²¹⁰ content	Remarks
	<u>picocurie/cigarette</u>	
Cigarettes	0.43	Results of Radford and Hund (ibid.)
Cigarettes	0.29 ± 0.03	two 10-cigarette batches
	<u>picocurie/gram</u>	
Pipe Tobacco "M"	0.27 ± 0.04	-
Pipe Tobacco "L"	0.30 ± 0.04	-

These results are not surprising in light of the report by Mayneord³ which gives the polonium-210 content of numerous samples of raw, dried herbiage as 1.0 to 20 picocuries per gram. Approximately equal amounts of lead-210 were found, indicating that the contamination arises from the natural decay of uranium-238 and radium-226

²E. P. Radford, Jr., and V. R. Hunt, Science, **143**, 3603 (1964).

³W. V. Mayneord, Nature, **184**, 667 (1959).

POLONIUM-209 HALF-LIFE

A review of the measurements to determine the half-life of polonium-209 indicated that a relatively rapid determination of the half-life could be made by measuring the ratio of the alpha activities of polonium-208 and polonium-209 as a function of time in a source containing both isotopes. The ratio of alpha activities can be expressed as:

$$R_{A_t} = \frac{R_{A_o} e^{-\lambda_8 t}}{e^{-\lambda_9 t}} \quad (1a)$$

or

$$R_{A_t} = R_{A_o} e^{-(\lambda_8 - \lambda_9)t} \quad (1b)$$

where R_{A_t} = the alpha activity ratio at any time t ,

R_{A_o} = the alpha activity ratio at time zero,

λ_8 = the decay constant of Po^{208}

λ_9 = the decay constant of Po^{209}

Equation (1b) indicates the improvement of the ratio method over the case of following total alpha activity, or a physical quantity related to total alpha activity, such as heat output. In the ratio method, only two unknowns, R_{A_o} and $(\lambda_8 - \lambda_9)$, are present. In the direct measurement of total alpha activity, an equation with four unknowns must be used for total activity as a function of time;

$$A_t = A_8^o e^{-\lambda_8 t} + A_9^o e^{-\lambda_9 t} \quad (2)$$

where A_t is the total activity at any time, t ,

A_8^o is the activity of Po^{208} at time zero,

A_9^o is the activity of Po^{209} at time zero,

and the remaining symbols are the same as equation (1b).

Data on the ratio of alpha activities of Po^{208} and Po^{209} have been accumulated over a period of one year and are shown in Figure 1, where R_{A_t} is plotted as a function of time. Data points were obtained with high-resolution alpha pulse-height analysis equipment, consisting of a semiconductor surface barrier detector, an ORTEC 101-201 amplifier preamplifier combination, and an RIDL 400-channel analyzer. A typical alpha spectrum is shown in Figure 2. The area under the alpha peak is determined by summing the counts in each channel in the peak, and the data were analyzed by a least-squares computer program.

The half-life of polonium-209 was found to be 42.4 ± 6.0 years, assuming that the polonium-208 half-life was 1055 ± 2 days. This value of 42.4 ± 6.0 years is in moderately good agreement with the half-life obtained by calorimetric measurements from a variational analysis fit of calorimetry data and that reported previously⁴ where a half-life of 40.2 ± 2.6 years was determined by spectrophotometric estimation of total polonium combined with gamma counting. The error in the half-life obtained by the ratio method should decrease as more data points are obtained.

⁴G. R. Grove, L. V. Jones and J. F. Eichelberger "Mound Laboratory Progress Report for December, 1963" MLM-1179.

Figure 1 The Alpha Activity Ratio Polonium-208 and Polonium-209 as a Function of Time in a Sample Containing Both Isotopes.

Figure 2 Alpha Pulse-Height Spectrum of Polonium-208 and Polonium-209.

DECAY SCHEME STUDIES

Calculations given in a previous report indicated that the electron-capture decay energy of polonium-208 might be sufficient to populate a 1.42 Mev level in bismuth-208. The 1.42 Mev level is metastable, with a half-life of 2.7×10^{-3} seconds.

A block diagram of the electronics equipment for the experimental detection of the delayed state is shown in Figure 3.

The equipment was checked out first using selenium-75, which decays by electron-capture to astatine-75, as shown in Figure 4.

Referring to the block diagram in Figure 3, the 17-millisecond state in astatine-75 was detected by amplifying all gamma-ray events detected in the well crystal sent to the two single channel analyzers. The "GATE" single-channel analyzer was set to produce a pulse when the gamma ray energy was 0.097 Mev (from the 0.402 - 0.305 transition). The occurrence of a gamma ray of this energy was taken as a signal that the 0.305 Mev level in astatine-75 was created. The pulse from the "GATE" single-channel analyzer was fed to the time-base generator, which then produced a series of channel-advance singles which were fed to the multichannel analyzer. The time-base generator was set to provide a sweep with a channel width of 800 microseconds over 100 channels. The "DETECTOR" single-channel analyzer was set to produce a pulse when a gamma-ray event of 0.280 Mev occurred. These pulses were sent to the multichannel analyzer, and were accepted only when the analyzer had been turned "on" by the train of channel-advance singles from the time-base generator. Because of the overlap of the 0.136-Mev gamma ray in the "GATE" single-channel analyzer and the 0.265-Mev gamma ray in the "DETECTOR" single-channel analyzer, background counting rates were high, requiring a large number of sweeps to be recorded before a reliable estimate of the half-life of the excited state was obtained.

Figure 3 Block Diagram of Equipment Used to Detect Millisecond Delayed States.

Figure 4 Decay Scheme of Se^{75}

The data collected after a total of 1,122,159 sweeps is shown in Figure 5, where the counts per channel is plotted as a function of time. For clarity and emphasis on the shape of the curve, 5,000 counts per channel were subtracted from each data point. A least-squares fit of the data by a variational method, in which the background was taken to be constant, gave a half-life for the 0.305-Mev level in astatine-75 as 15.64 ± 0.92 milliseconds, in good agreement with the literature value⁵ of 17.0 ± 1.5 milliseconds.

A similar experiment was undertaken to determine the half-life of the metastable state at 1.42 Mev in bismuth-208 which may be populated in the electron-capture decay of polonium-208. Again referring to Figure 3 the "GATE" detector in this case was set to provide a pulse to the time base generator when an x-ray event of 0.075 Mev occurred. A photon of this energy is emitted when polonium-208 decays by K-electron capture. Because the half-life of the bismuth-208 level at 1.42 Mev has been given as about three milliseconds, a higher sweep rate was required. In this case, the time-base generator was set to provide a sweep with a channel width of 100 microseconds over 100 channels. The "DETECTOR" single-channel analyzer was set to provide a pulse to the multichannel analyzer when a gamma-ray event with an energy between 0.870 to 0.970 Mev was obtained. This wide window was chosen because of the uncertainty in the gamma-ray energy involved in the delayed-state decay reported by Vegors and Axel.⁶ The background counting rates were expected to be higher in the bismuth-208 run than in the astatine-75 experiment, because the 0.75-Mev K-x-ray occurs each time polonium-208 decays by electron capture to any level in bismuth-208, and some K-x-rays are excited in the alpha decay of polonium-208. Thus, the number of sweeps initiated by electron-capture decay to the 1.42 Mev level in bismuth-208 would be only a small fraction of the total sweeps.

⁵A. W. Schardt, *Phys. Rev.* **108**, 298 (1957).

⁶S. H. Vegors, Jr., and P. Axel, *Phys. Rev.* **101**, 1067 (1957).

Figure 5 Counting Data as a Function of Time in the Measurement of the Half-life of the 0.305 Mev Level in As^{75} .

A total of 5,393,204 sweeps were accumulated over a period of seven days. No significant curvature was observed in the data. An attempt will be made to obtain a computer analysis of the data; and if these results are encouraging, another experiment will be performed in which more sweeps will be accumulated. Calculations are in progress to determine limits of detectability for the experiments.

Po²⁰⁸-Po²⁰⁹ Studies The alpha-gamma coincidence spectrum in Po²⁰⁸ and Po²⁰⁹ is being determined. The measurement system consists of gold silicon surface barrier detectors, 2 x 2 inch NaI(Tl) scintillation detectors, a fast-slow coincidence circuit, and a multi-channel analyzer. Net coincidence count rates obtained by gating on all the gamma rays above 40 kev is about 0.5 counts per minute, thus requiring counting times of several thousand minutes, even with modest intensity peaks.

It appears that two alpha groups of 4.62 and 4.78 Mev are in coincidence with the gamma rays. The intensity of the 4.78 Mev group on the spectrum obtained was only 1/30 of the 4.62 Mev group; hence, its presence is not firmly established.

RESIDUE ADSORPTION

Difficulty is still being experienced in preparing a barium-140 stock solution free of calcium. The solutions resulting from purifications made by cation exchange, by residue adsorption, or by radiocolloidal filtrations appear at first to be carrier-free when evaporated to dryness, and initial test separations produce the expected results, i.e., approximately 90 per cent of the barium-140 is desorbed while leaving behind approximately 90 per cent of the lanthanum-140 adsorbed as the hydroxide. However, after a period of days, the separations become erratic, and the residues no longer appear carrier-free.

It seems possible that there is radiolytic attack on the freshly cleaned Pyrex glass bottles which are used for storing the stock solutions and that the calcium impurity is reintroduced by the storage vessel. If this proves to be the case, storage in polyethylene vessels should eliminate this problem.

HALF-LIFE OF RADIUM-223

Computation of the half-life of radium-223 and consolidation of the data from alpha-counting and calorimetry measurements are nearly completed. The data from the 10 samples which were alpha-counted have been treated by the method of decaying pairs (MLM-1128, page 11) and by the variational method (MLM-1151, page 9). To eliminate the uncertainty in the nature of the function which describes resolution time losses in the proportional counter (MLM-1175, page 9), only counting rates below 8×10^5 counts per minute were used in the computations. In all cases, the data were weighted inversely as the squares of their probable errors, and Chauvenet's criterion was employed for the rejection of inconsistent points.

In Table 3, the results are shown for the half-life of radium-223 computed by the decaying pair method from ten samples taken two at a time. Table 4 lists the results obtained from the same ten samples by the variational method.

It is seen in Table 3 that, of the four pairs rejected by Chauvenet's criterion in the computation of the weighted grand mean, three included sample number 9. Furthermore, Birge's test for consistency yielded a value of h_x greater than 1.86, indicating that the means were inconsistent.

However, when the grand mean was recomputed, with sample number 9 eliminated, only pair 1-3 was rejected by Chauvenet's criterion, and the value of h_x decreased to 0.82, well within the prescribed limit. (The rejection of pair 1-3 has already been discussed. (See MLM-1148, Table 12, page 22).

In Table 4, three treatments of the variational method have been applied. In two cases, the resolution time was varied to yield the best value of the half-life: τ_1 was varied in the Ruark and Brammer equation, and

Table 3

HALF-LIFE OF RADIUM-223 BY DECAYING PAIRS

(Count rate limit: 8×10^5 c/m; Data Weighted;
Chauvenet's Criterion Used)

Sample Number	Half-life (days)	Prob. Error (days)	hx ^a	Rejected Pairs
1	11.4296	0.0053	1.10	1-3, 1-9
2	11.4325	0.0052	0.51	2-9
3	11.4350	0.0057	0.54	3-1, 3-9
4	11.4288	0.0042	0.35	4-9
5	11.4264	0.0057	2.79	none
6	11.4382	0.0030	1.49	6-7, 6-8, 6-9
7	11.4480	0.0037	1.34	none
8	11.4504	0.0057	0.91	none
9	11.4073	0.0118	7.78	none
10	11.4332	0.0038	0.16	none
WGM (45) ^b	11.4331	0.0024	2.74	1-3, 1-9, 3-9, 4-9
WGM (90) ^c	11.4381	0.0014	2.12	none
WGM (36) ^d	11.4374	0.0022	0.82	1-3

^aBirge's criterion of consistent means (MLM-1179).^bWeighted grand mean for 45 pairs.^cWeighted grand mean of 10 samples, each comprising 9 pairs.^dWeighted grand mean for 36 pairs; sample number 9 eliminated.

r_2 in the Volz equation (MLM-1179, page 10). There was no significant difference between the half-life values computed by the two equations, either for individual samples or for the weighted grand means. It will be noted, however, that the value of hx in each case was quite high.

In the last column of Table 4, the results of varying the half-life are shown. In this case, while there is no significant difference in the computed half-life, for each sample, the probable errors have all been increased by an order of magnitude. The explanation for this difference lies in the method of calculating the probable error; in the first two cases, the value of r used in the final computation of the half-life is treated as exact, with no uncertainty. In the last case, the probable error is computed from the parabola generated by varying the half-life, and the probable error in the resolution time becomes a component of the probable error in the half-life. The details and justification for this method of computation will be discussed in a later report. It is immediately apparent, however, from the value of hx , that the increase in the orders of magnitude of the probable errors has the effect of making the weighted grand mean more consistent, without significantly changing the computed half-life value. At the same time, the probable error in the grand mean is increased by only a factor of two.

Table 4

HALF-LIFE OF RADIUM-223 BY VARIATIONAL METHOD

(Count rate limit: 8×10^5 c/m; Data Weighted;
Chauvenet's Criterion Used)

Sample Number	Half-life (Days) (r_1 varied)	Half-life (Days) (r_2 varied)	Half-life (Days) ($T_{1/2}$ varied)
1	11.4115 ± 0.0025	11.4156 ± 0.0025	11.4110 ± 0.0210
2	11.4351 ± 0.0025	11.4407 ± 0.0025	11.4343 ± 0.0217
3	11.3998 ± 0.0039	11.4031 ± 0.0039	11.3973 ± 0.0239
4	11.4278 ± 0.0019	11.4338 ± 0.0019	11.4272 ± 0.0199
5	11.4160 ± 0.0024	11.4241 ± 0.0024	11.4107 ± 0.0183
6	11.4495 ± 0.0022	11.4368 ± 0.0023	11.4487 ± 0.0175
7	11.4452 ± 0.0022	11.4400 ± 0.0023	11.4492 ± 0.0217
8	11.4479 ± 0.0025	11.4520 ± 0.0024	11.4238 ± 0.0186
9	11.4356 ± 0.0034	11.4375 ± 0.0035	11.4336 ± 0.0194
10	11.4189 ± 0.0017	11.4165 ± 0.0020	11.4181 ± 0.0183
WGM ^a	11.4296 ± 0.0032	11.4311 ± 0.0028	11.4267 ± 0.0062
h_x ^b	10.69	8.48	1.42

^aWeighted grand mean for 10 samples.^bBirge's criterion of consistent means (MLM-1179)-

The good agreement between the calorimetric and alpha-counting results is shown below:

Calorimeter 67	11.4344 ± 0.0011
Calorimeter 60	11.4432 ± 0.0057
Alpha-Counting (Decaying Pairs)	11.4374 ± 0.0022
Alpha-Counting (Variational)	11.4267 ± 0.0062

VISCOSITY OF PLUTONIUM ALLOY

As part of the liquid metals studies of potential plutonium reactor fuels, the viscosity of a molten 99 weight per cent plutonium-one weight per cent gallium alloy was measured at 12 temperatures from its 669°C liquidus temperature to 816°C by an absolute method in an oscillating cup viscosimeter. In this type of apparatus, the viscosity is a function of the period of oscillation and damping effect of the liquid metal upon the normal oscillation of the torsion pendulum formed by suspending the tantalum viscosity capsule on a torsion fiber.

The viscosity decreased linearly when viscosity (η in poise) was plotted as $\text{Log } (\eta/T)$ as a function of reciprocal absolute temperature ($1/T$). The viscosity data were fitted to a straight line (Figure 5) by a least squares treatment. The equation for the line is:

$$\text{Log } \left(\frac{\eta}{T} \times 10^5 \right) = 1.3879 \frac{1000}{T} - 0.4998$$

where

$$\sigma_{\eta} = 0.0015 \text{ poise (by Standard Deviation)}$$

$$E = 6351 \text{ cal/g-atom (Activation Energy)}$$

The addition of one weight per cent gallium to plutonium resulted in a 50 per cent increase in viscosity as compared to unalloyed plutonium over the same temperature interval. The measured viscosity coefficients at the twelve temperatures investigated are recorded in Table 5. The viscosities calculated from the straight line representing a least squares fit of the experimental data are shown in Table 6.

The liquidus temperature for this alloy was determined with the viscosimeter. When the temperature of the alloy was decreased below the liquidus temperature, the appearance of the first solid precipitate in the melt caused an abrupt increase in viscosity. This increase in viscosity caused a correspondingly abrupt increase in the period of oscillation. When the period of oscillation was plotted as a linear function of temperature, two sets of data points emerged. In this case, three measurements were made in the liquid plus solid region of the binary system and 13 observations were made in the liquid state. By drawing straight lines through these two sets of data points, the liquidus temperature of $669 \pm 2^{\circ}\text{C}$ was defined. (See Figure 6).

Estimated liquid density values were used in the calculations of the viscosities in this series of experiments although the experimental determinations of density are planned. Since a one per cent error in liquid density results in a corresponding one per cent error in the calculated viscosity, the reported viscosities are subject to revision when precise liquid densities for this alloy are measured.

Figure 6 Viscosity of a Pu-1% Ga Alloy as a Function of Reciprocal Absolute Temperature.

Table 5
MEASURED VISCOSITY COEFFICIENTS

Temperature °C	Density ^a gm/cc	Viscosity Centipoise
816	16.01	6.34
783	16.08	7.12
771	16.09	6.88
757	16.10	7.25
732	16.16	7.90
725	16.17	7.67
710	16.19	8.12
699	16.20	8.12
689	16.21	8.38
684	16.21	8.70
677	16.23	8.50
672	16.23	8.72

^aEstimated from atomic volumes of alloy constituents.

Table 6
CALCULATED VISCOSITY COEFFICIENTS^a

Temperature °C	Viscosity Centipoise
815	6.60
775	7.07
725	7.79
669	8.82

^aViscosities are calculated from a line describing the least squares fit of the experimental data on a plot of $\text{Log } \eta/T$ as a function of $1/T$.

ALPHA AND NEUTRON SOURCES

Mound Laboratory is responsible for producing alpha and neutron sources, manufactured from polonium-210 and plutonium-239, which cannot be produced by American industry at the present time. The techniques of fabricating these unusual sources are being developed and publicized.

NEUTRON MEASUREMENTS

The increase of neutron emission in PuBe sources due to the ingrowth of americium-241 is being measured by calorimetric techniques, and by neutron measurements using the precision long counter as the detector. The neutron emission is being measured using neutron sources manufactured from the americium-241, plutonium-239 and plutonium-238.

Preliminary results indicate that the neutron emission of the americium-241 source is decreasing about 0.4% per year faster than expected from both the predicted and measured decay of americium-241. Alpha spectra analysis and calorimeter measurements indicate no other isotope is present with the americium-241 in sufficient quantities to account for this relatively large decrease in neutron emission. One possible explanation for this relatively fast decrease in neutron emission is a slow, uniform separation of the americium and beryllium in this source. These two elements were not alloyed in this source during its fabrication.

Until it was realized that the americium-beryllium source was not as stable as had been expected, it had had been used extensively in measurements as a comparison standard. It has now been discarded in favor of a plutonium-beryllium source, whose ingrowth of americium-241 is thought to be less than about 0.1% per year.

ISOTOPE SEPARATION

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.

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 to the thermal diffusion separation systems, and the evaluation of various other separation methods, such as chemical exchange and distillation.

The carbon-13 fraction was separated from natural methane. As of February 11, 1964, total production from the 11-column thermal diffusion cascade totaled approximately 0.85 g of carbon-13. A composition profile for the 11-column cascade was measured on February 5, 1964. The data are shown in Table 7 and they have not been integrated yet to get total carbon-13 holdup; spot comparisons of mass-17 enrichment show clearly that no depletion of the system inventory of carbon-13 has taken place since December 27, the date of the last profile measurement. This means that a carbon-13 production rate can be sustained on a long term basis at 0.6 gram per month.

Measurement of Carbon Monoxide Transport Coefficients Thermal diffusion transport measurements were completed for carbon monoxide in a 3/4-inch diameter column with a 1/16-inch diameter center wire. The transport coefficients H' , K_c' , and K_d were measured at 350° and 500°C for both mass 29 and mass 30. The transport coefficients are defined by the differential equation for isotope transport in a thermal diffusion column:

$$\tau = H' p^2 c (1-c) - (K_c' p^4 + K_d) \frac{dc}{dz}$$

where τ is the rate of transport of the desired isotope toward the positive end of the column, c is the fraction of the desired isotope, p is the pressure, and z is distance along the column.

Experimental results are given in Table 8 along with theoretical values derived from the shape factor tabulation of McInteer and Reisfeld.⁷

Agreement between measured and predicted transport coefficients is close for mass 29 carbon monoxide. Agreement is not as close for mass 30 carbon monoxide, probably because of the poor precision in the determination of isotope ratios for this species. The combination of low mass 30 abundance and the sensitivity limitations of the presently available mass spectrometer gives an expected $\pm 10\%$ error in isotope ratio measurements at either end of the experimental thermal diffusion column. This leads to large errors in transport coefficients derived from the isotope ratios.

⁷B. B. McInteer and M. J. Reisfeld, LAMS 2517, February, 1961.

Table 7

COMPOSITION PROFILES FOR THE 11-COLUMN METHANE THERMAL DIFFUSION CASCADE

Sample Point	December 27, 1963		February 5, 1964	
	Methane Purity (%)	Mass-17 Enrichment (%)	Methane Purity (%)	Mass-17 Enrichment (%)
Waste Stream	99.60	0.96	99.53	0.96
Feed	99.62	1.09	—	1.09
Stage 1 Bottom	99.32	1.31	99.49	1.38
Stage 2 Bottom	99.21	2.45	99.37	2.85
Stage 3 Bottom	99.32	3.82	99.32	4.26
Stage 4 Bottom	99.18	6.93	99.20	7.25
Stage 5 Bottom	—	—	98.21	19.10
Stage 6 Bottom	97.50	48.90	98.42	50.46
Stage 7 Bottom	97.46	83.31	95.81	93.04
Stage 8 Bottom	91.10	97.24	89.98	95.98
(Product Withdrawal Point)				
Stage 9 Middle	55.92	97.95	64.12	97.06

Table 8

THERMAL DIFFUSION COLUMN TRANSPORT COEFFICIENTS FOR CARBON MONOXIDE

Mass 29 CO	350°C	500°C
H' (exp)	2.48×10^{-5}	3.68×10^{-5} g/sec-atm ²
H' (theory)	2.83×10^{-5}	3.97×10^{-5}
K'_c (exp)	5.53×10^{-2}	5.22×10^{-2} g/cm-sec-atm ⁴
K'_c (theory)	4.87×10^{-2}	4.26×10^{-2}
K_d (exp)	7.17×10^{-4}	8.64×10^{-4} g/cm-sec
K_d (theory)	7.75×10^{-4}	8.36×10^{-4}
Mass 30 CO		
H' (exp)	3.93×10^{-5}	6.60×10^{-5} g/sec-atm ²
H' (theory)	5.66×10^{-5}	7.94×10^{-5}
K'_c (exp)	4.87×10^{-2}	6.24×10^{-2} g/cm-sec-atm ⁴
K'_c (theory)	4.87×10^{-2}	4.26×10^{-2}
K_d (exp)	10.85×10^{-4}	8.74×10^{-4} g/cm-sec
K_d (theory)	7.75×10^{-4}	8.36×10^{-4}

HELIUM-3 VAPOR PRESSURE THERMOMETRY

As part of an effort to establish the vapor pressure of helium-3 as a temperature scale below 3.2°K, the thermomolecular pressure drop in a tube possessing a temperature gradient is being determined. This pressure drop will be determined as a function of temperature and tube diameter in order that it may be applied as a correction to vapor pressure measurements in low temperature thermometry.

Helium-3 is significantly more useful than helium-4 as the working fluid in a vapor pressure thermometer for the following reasons:

1. The light isotope has a higher vapor pressure between 0.3° and 3.2°K.
2. Helium-3 vapor pressure possesses a larger temperature dependence than helium-4, and is thus more sensitive as a thermometer.
3. Helium-3 does not have the troublesome superfluid properties as does helium-4.

To eliminate heat leaks, detailed calculations were made on the present helium thermometer system which contains 1/4-inch and 1/32-inch tubes, and for which some estimate of the total heat leak is available from past experiments. Of the total calculated heat leak, about 70% was attributed to thermal conduction, from ambient down the 1/4-inch and 1/32-inch thermometer tubes to the helium vapor pressure reservoir. In the design of the next thermometer system, thermal grounding of the vapor pressure tubes to the helium-4 bath can greatly reduce the heat leak into the thermometer bulb.

THERMAL DIFFUSION THEORY

The thermal diffusion column, which was introduced in 1938 by Clusius and Dickel, has proven to be a convenient method of separating gaseous isotopic species. For example, at Mound Laboratory they are used to separate the isotopes of the inert gases and $C^{13}H_4$ and $C^{12}H_4$. Soon after the introduction of the thermal diffusion column, Jones and Furry developed a theory to explain the performance of a column; however, the quantitative agreement between this theory and experimental results have generally been poor. Although the existence of this poor quantitative agreement has been known for several years, a satisfactory explanation for it has never been given. Since thermal diffusion columns are used extensively at Mound Laboratory, an attempt is being made to determine the reasons for these discrepancies.

An equation was derived recently⁸ for the transport of species one up the column. This equation contains several integrals which are implicit functions of the concentration. The presence of the concentration in these integrals makes the equation extremely difficult to treat, so it is desirable to try to remove this dependence from the integrals. In order to do this, the concentration dependence of the function

$$G(r, c_1) \equiv \int_{r_2}^r m_1 n v(r, c_1) r dr$$

must be determined.

The differential equation for the convective velocity, $v(r, c_1)$, is

$$\frac{1}{r} \frac{d}{dr} \eta r \frac{dv}{dr} = \frac{dp}{dz} + \rho g.$$

or

$$\frac{d}{dr} \frac{1}{r} \frac{d}{dr} \eta r \frac{dv}{dr} = g \frac{d}{dr} \rho$$

⁸G. R. Grove, L. V. Jones and J. F. Eichelberger "Mound Laboratory Progress Report for December, 1963, " MLM-1179.

Recently, it was shown⁹ that for isotopic mixtures, the coefficient of viscosity, η , is a linear function of the concentration, i.e.,

$$\eta(c_1) = \left[1 - \frac{M_2^{1/2} - M_1^{1/2}}{M_1^{1/2}} c_1 \right] \eta_2$$

where the subscripts refer to species 1 and species 2. Also,

$$\rho(c_1) = \left[1 - \frac{M_2 - M_1}{M_2} c_1 \right] \rho_2$$

Substituting these expressions for $\eta(c_1)$ and $\rho(c_1)$ into the differential equation for $v(r, c_1)$ and neglecting the variation of c_1 in the radial direction, one obtains the differential equation

$$\frac{d}{dr} \frac{1}{r} \frac{d}{dr} r \eta_2 \frac{d}{dr} \mu v = g \frac{d}{dr} \rho_2$$

where

$$\mu = \frac{\left[1 - \frac{M_2^{1/2} - M_1^{1/2}}{M_1^{1/2}} c_1 \right]}{\left[1 - \frac{M_2 - M_1}{M_2} c_1 \right]}$$

Since μ and v are the only quantities in this equation which are functions of c_1 , μv must be independent of c_1 . Letting $v_2(r) = \mu v$, it is seen that this is the differential equation for the convective velocity for a pure gas of species 2. Thus,

$$v(r, c_1) = \frac{1}{\mu(c_1)} v_2(r)$$

Expanding $1/\mu$ in a power series about $M_1 = M_2$, this equation can be written

$$v(r, c_1) = \left[1 - \frac{(M_2 - M_1)}{2M_1} c_1 + \frac{(M_2 - M_1)^2}{8M_2^2} c_1(1 - 2c_1) \right] v_2(r)$$

or

$$v(r, c_1) = g(c_1) v_2(r)$$

For most applications, the third term in the above equation can be ignored.

Using the above expression for $v(r, c_1)$, $G(r, c_1)$ can be written

$$\begin{aligned} G(r, c_1) &= g(c_1) \int_{r_2}^r m_1 n v_2(r) r dr \\ &= g(c_1) F(r) \end{aligned}$$

With this expression for $G(r, c_1)$, the concentration can be factored out of the integrals which were given previously.⁹

⁹G. R. Grove, L. V. Jones and J. F. Eichelberger, "Mound Laboratory Progress Report for January, 1964", MLM-1183.

The quantities η , ρ , D , and α are functions of the temperature. The relation between T and r is given by the equation for the temperature gradient, i.e.,

$$2\pi Q = 2\pi r \lambda \frac{\partial T}{\partial r}$$

where Q is the heat flow by conduction per unit length of column. Q is determined by integrating this equation from r_2 to r_1 , thus

$$Q = \frac{1}{\ln(r_1/r_2)} \int_{T_1}^{T_2} \lambda(T, c_1) c T$$

The thermal conductivity can be written as

$$\lambda(T, c_1) = F(c_1) \lambda_2(T)$$

where $\lambda_2(T)$ is the thermal conductivity for species 2. Using these expressions for λ and Q , the equation for the temperature gradient is

$$\frac{dT}{dr} = - \frac{1}{r \lambda_2 \ln(r_1/r_2)} \int_{T_1}^{T_2} \lambda_2 dT$$

Thus, the temperature gradient is independent of concentration and no concentration dependence is introduced into the equations in transforming from r to T .

PLASTICS

The physical and chemical properties of various plastic formulations are being determined for specialized applications.

Foams Evaluation Various foamed formulations were prepared, as shown in Table 9, and the resistances of these foams to heat and solvents were determined.

Table 9

EPOXY FOAM FORMULATIONS

Sample	Epon 828	Oxiron 2001	Diaminodiphenyl Sulfone	Curing Agent U	Trimethoxy- Boroxine	Cab-o- Sil	Acetone	DC-113
EF 1	20 g (100) ^a	—	8 g (40)	—	12 g (60)	0.5 g (2.5)	1.0 g (5)	1.0 g (5)
EF 2	20 g (100)	—	—	14 g (70)	4 g (20)	0.5 g (2.5)	1.0 g (5)	1.0 g (5)
EF 3	12 g (60)	8 g (40)	8 g (40)	—	12 g (60)	0.5 g (2.5)	1.0 g (5)	1.0 g (5)
EF 4	12 g (60)	8 g (40)	—	14 g (70)	4 g (20)	0.5 g (2.5)	1.0 g (5)	1.0 g (5)

^aNumbers in parenthesis refer to parts per hundred resin. The gram quantities are for a 1X batch.

To prepare samples of formulations EF1 and EF3, the ingredients, less trimethoxy-boroxine (TMB), were mixed into a master batch and stirred thoroughly for ten minutes. A 30.5-gram aliquot was placed in a three-ounce untreated paper cup, and 12 grams of TMB were added. The mixture was stirred by hand and placed in a closed cardboard mold (1.5 x 1.5 x 1.5 inch) with a small vent hole in the lid. This type of mold was used to restrict the expansion of the foam (to partially control the cell structure) and to simulate proposed application conditions. Because of the extremely low cream time (< 1 minute), some difficulty was encountered in transferring the EF3 mixtures to the cardboard mold.

Attempts to use the master batch technique for formulations EF2 and EF4 were unsuccessful, possibly because increased mass of curing agent and resin caused rapid gelation of the mixture. Samples of EF2 and EF4 were prepared by mixing the components, less TMB, in a three-ounce paper cup, adding TMB, and transferring the mixture to a cardboard mold.

The exotherm generated during foaming was measured with an iron-constantan thermocouple. Exotherms for EF1, EF2, and EF3 were 110°C, 132°C, and 136°C, respectively. All samples were postcured at 70°C for 24 hours.

The foam samples were cut into 1.2-inch cubes for density (ASTM 1622-59T) and compressive strength (ASTM 1621-59T) determinations. The large variations in density of the samples from any one formulation were observed and were probably a result of the inability of the cardboard molds to effectively restrict foaming. Since compressive strength is a function of sample density, a similar lack of precision was noted in the values obtained for compressive strength. Thus, the results listed in Table 10 indicate only the range of density and compressive strength that result from these formulations.

Table 10
PROPERTIES OF EPOXY FOAMS

Sample	Density (lb/ft ³)	Compressive Strength (psi)	% Compression	Solvent Resistance (%wt/loss)				Heat Resistance (%wt/loss) ^b		
				Benzene	Ethanol	Acetic Acid	Dimethyl Formamide	260°C	300°C	340°C
EF 1	11	29	3.5 ^a	4.4	53.2	16.0	88.6	15.8 (R) ^c	21.7 (R)	34.9 (d)
EF 2	21	680	5.3	0.2	10.8	8.5	12.4	18.1 (SR)	37.3 (SR)	59.2 (d)
EF 3	13	180	5.0	6.1	49.5	39.5	47.6	14.9 (R)	19.8 (R)	28.2 (R)
EF 4	31	597	3.0	6.2	40.0	24.9	10.9	12.2 (SR)	35.2 (S)	61.5 (d)

^aIndicated compression at maximum pressure.

^bBased on original sample weight.

^cLetters in parenthesis refer to rigidity of sample before removal from oven when R- rigid; SR- Semi-rigid; S- soft; and d- decomposed.

The resistance of the foams to selected solvents was determined by placing weighed samples (1/4 x 1/4 x 3/4 inch) in weighing battles with various solvents. The samples were immersed for seven days, the solvent was decanted off, and the samples were exposed to the atmosphere for an additional seven days. The samples were reweighed to determine the weight loss.

The effect of short term exposure to high temperature was determined by placing a weighed sample (1 x 1 x 1/4 inch) in an oven for one hour. The sample was examined for rigidity while still in the oven. After cooling to room temperature, the percent weight loss was determined.

The compressive strength of the epoxy foams, with the exception of EF2, were low compared with values reported in the literature. The results obtained for EF2 were in relatively good agreement with reported compressive strengths. The brittleness of formulations EF1 and EF4 might explain their low compressive strengths.

Of the four formulations, EF2 had the best solvent resistance. It was scarcely affected by benzene and only moderately affected by other solvents. However, the general solvent resistance of all the foams was low. Formulation EF3 was the most heat resistant formulation. (See Table 10). Samples of this formulation remained rigid at all test temperatures. Below 260°C, formulation EF2 possessed superior properties. For temperature application, however, formulation EF3 is superior.

ADHESIVES

Adhesives are being developed for use with explosives and for other specialized applications. The adhesives must have satisfactory working and curing characteristics; they must retain flexibility at low temperatures and adequate tensile strength at elevated temperatures.

Three manganese dioxide-cured, polysulfide-base conductive adhesives were formulated and evaluated (Table 11). These adhesives are more flexible than the corresponding lead peroxide-cured materials pre-

viously reported.¹⁰ The formulation with the highest silver content exhibited the lowest electrical resistivity. Formulation CS-31 is adequate in all respects except for its low peel strength.

Table 11

COMPOSITION AND PROPERTIES OF POLYSULFIDE CONDUCTIVE ADHESIVES^a

Component	Component in Formulation (wt %)		
	CS-25	CS-29	CS-31
LP-33 Prepolymer ^b	19.5	18.3	16.8
Activated Manganese Dioxide ^b	1.1	1.1	1.0
Activator Solution ^c	3.3	2.6	2.4
Continex CF Black ^d	5.5	5.6	4.4
J-2 Silver Powder ^e	70.6	72.4	75.4
Electrical Resistivity (ohm-cm)			
A. Aged 1 wk at room temperature after cure	0.63	0.15	0.05
B. As in A + 24 hrs at 160°F and cooling	0.60	0.31	0.06
Tensile Bond Strength (psi)	280	300	280
Peel Strength (lbs/in.)	7	7	6
Ultimate Elongation of Film Strip at -65°F (%)	-	11	10

^aAll formulations had a working pot life of more than 2 hrs. All specimens cured 24 hrs at 160°F.

^bThiokol Chemical Corp.

^cA mixture of 16.6 wt % each of benzene, methyl dinitrobenzene, and dibutyl phthalate and 50.2% Aroclor 1254 (Monsanto Chemical Co.).

^dWitco Chemical Co.

^eEngelhard Ind.

Substitution of prepolymer LP-3 for LP-33 in the above formulations did not appreciably change their handling characteristics.

The commercial unfilled silicone potting material LTV-602 (General Electric) cured by a basic catalyst was reinvestigated as a base for conductive adhesives (Table 12). All formulations exhibited poor peel strength with relatively high resistivities. The resistivities increased on aging at room temperature. However, since these formulations show great flexibility at low temperatures, they may be suitable for some applications and will be investigated further.

¹⁰G. R. Grove, L. V. Jones and J. F. Eichelberger, "Mound Laboratory Progress Report for December, 1963", MLM-1179

Table 12

COMPOSITION AND PROPERTIES OF SILICONE BASE ADHESIVES^a

Component	Component in Formulation (wt %)				
	SL-1	SL-2	SL-3	SL-4	SL-5
Catalyzed LTV-602 Silicone ^b	22.5	77.0	20.0	20.0	20.0
Silver Flake MD-750 ^c	77.5	—	—	—	—
Silver Powder J-2 ^d	—	—	80.0	79.5	77.0
XC-3001 Carbon Black ^e	—	23.0	—	0.5	3.0
Electrical Resistivity (ohm-cm) 1 day after cure	>1000	>1000	0.10	0.61	63
1 wk after cure	>1000	>1000	0.42	3.78	151
2 wks after cure	>1000	>1000	0.53	3.78	292
Ultimate Elongation of Film Strip at -65°F (%)	—	185	Samples Failed	73	67
Peel Strength (lbs/in.)	0.7	0.5	0.2	0.3	0.5

^aAll formulations were cured 16 hrs at 160°F.

^bLTV-602, 99.5 wt % and SRC-05, 0.5 wt % (General Electric Co.).

^cMetals Disintegrating Division, Martin-Marietta Corp.

^dEngelhard Ind.

^eUnion Carbide Co.

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.

CALORIMETRY

Twin bridge Calorimeter 91 was constructed and tested. It is similar to Calorimeter 39 and almost identical Calorimeter 90.¹¹ Several modifications were made that should add significantly to its performance. Brass pump cylinder jacket stock was used for the jackets and a new baffle was developed. Tapered brass sleeves were used to bind the upper end of the thermels to the jacket. In general, more precision and care were exercised in all phases of the fabrication. A comparison of the sensitivities of the two sides is the best single test of the precision of construction.

The sensitivity of the sample side is 31616 microvolts per watt at five milliamperes of bridge current and the sensitivity of the dummy side is less by only 0.02%. The most probable difference would be about 0.1%, or a thickness of air gap within 0.0001 inch. A second test, which is not as sensitive, is the comparison of power of a sample that is assayed in both sides. An average of three runs with the sample side compared to a single run with the dummy side was within 0.0025%. The average of the four assays is within 0.003% of many assays on the same sample with Calorimeter 90.

A third test was made to observe the change in bridge potential when current was supplied to the heater leads only, and to compare this to the change in bridge potential when the same current is set in the heater. There was current in two leads on both sides of the calorimeter in each case. The relative bridge unbalance was 0.002%. Since the heat in the leads was equal to 0.5% of the heat in the heater, the leads were similar, one side to the other, to within 0.4%.

The bore of the brass pump cylinder stock is more precise than the jackets of Calorimeter 90, which were bored, or in other calorimeter jackets which were reamed. The jackets are 18.5 inches long, 1.5 inches ID and two inches OD. To minimize temperature interactions with the room, the upper four inches of the jackets were turned down to a wall thickness of 0.050 inch. Since the wall thickness of the jackets is 0.25 inch instead of the usual 0.125 inch for other calorimeters, the bath "noise" should be filtered more effectively.

The baffles, top and bottom, were made of Kel-F and aluminum. The baffle consists of a thin Kel-F shell. Discs of aluminum supported on thin Kel-F rings are in the shell which is 4.2 inches long, 1.120 inches OD, and 1.040 inches ID. The aluminum discs are 1.040 inches OD by 0.012 inch thick, and are spaced at intervals of 0.25 inch along the length inside the shell by 0.015-inch thick spacers of Kel-F. A cap, 0.25-inch thick, was bonded to the upper end of the shell with epoxy resin after this portion of the shell was chemically treated with Chemgrip. The lower edge of the top baffle shell is threaded into the top of the sample container.

Baffles of design similar to these have been made to replace the original baffles in Calorimeter 90 which were filled with Perlite and in Calorimeter 58 which were filled with Santocell. The sensitivity of Calorimeter 90 was increased by 0.01% and the sensitivity of Calorimeter 58 was decreased by 0.15%. The thermal resistance of the new baffles is between that of Perlite and Santocel filled baffles as might be expected. Santocel is a better insulator than air, and since the aluminum discs have a dull finish, the new baffles should have a thermal resistance less than that of air. The bottom baffles were similar to the top baffles except that the shell is also the form on which the heater leads are wound.

¹¹G. R. Grove, L. V. Jones and J. F. Eichelberger, "Mound Laboratory Progress Report for March, 1963", MLM-1152.

In other calorimeters, the upper end of the thermels have not fitted the jackets as tightly as desired. This was done so the thermels would not be racked when inserted into the jackets. In this calorimeter, the upper ends were made to fit the jacket loosely and, after insertion, a split brass sleeve with a slight taper was wedged between the end of the thermel and the jacket.

This calorimeter will be used primarily for half-life determinations. An automatic sample loading system will be installed which is especially helpful in half-life measurements.