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The Mound Laboratory Progress Report, issued monthly, is intended to be a means of reporting items of current technical interest in research and development programs. To issue this report as soon as possible after the end of the month, editorial work is limited; and since this is an informal progress report, the results and data presented are preliminary and subject to change.

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.

MONSANTO RESEARCH CORPORATION

A SUBSIDIARY OF MONSANTO CHEMICAL COMPANY

MOUND LABORATORY

MIAMISBURG, OHIO

OPERATED FOR

UNITED STATES ATOMIC ENERGY COMMISSION

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DISCLAIMER

TABLE OF CONTENTS

PLASTICS RESEARCH

Work is being directed at the development of methods of producing plastics and adhesives having characteristics superior to those currently being used at Mound Laboratory and elsewhere. Variations in plastic formulations and molding procedures are being studied in an effort to produce plastic which consistently meet specifications.

PLASTICS During the past month a satisfactory procedure for producing unfilled material was developed using production equipment. Previous materials gave about 10 to 15 per cent efficiency to Production while this material gives over 50 per cent molded parts for assembly. The size of the batch increased from 500 grams to 15 pounds, which represents a considerable advantage for production purposes. The standard formulation used was:

100	parts Dapon* 35	15 pounds
2	parts ter-butyl perbenzoate	136 grams
0.8	part benzoyl peroxide	54 grams
0.5	part 10-undecenoic acid	34 grams
	minimum amount of acetone	1½ to 2 pounds

To date 10 batches or 150 pounds have been made for production use.

The first 24 batches of Dacron+-filled diallyl phthalate were produced by research personnel to develop techniques for milling and handling this material. The remaining batches were produced by production personnel. A total of 76 batches were produced.

The plastics research group was asked to evaluate the material. Tensile strength, impact resistance, residual volatile content, cup flow time and cup weight tests were run.

Cup flow data, residual volatiles and cup weights were determined on each sample. Because of the time involved in molding test specimens, tensile and impact specimens were molded on every third batch.

Cup weights were taken on batches W-25 through W-76 to the nearest 0.01 gram. Since the volume of each cup is approximately equal, the cup weight gives a rough indication of density variations between batches. Cup weights varied between 30.39 grams and 31.00 grams. Overall average cup weight was 30.61 grams.

The range of tensile strengths was 3450 to 5478 psi. The average tensile strength was 4330 psi.

Izod impact resistance values varied from 2.54 to 5.25 ft. lbs. per inch notch: average was 4.11. Sixty per cent of the samples tested had average impact strengths in excess of 4.00 ft. lbs. per inch notch.

There was considerable variation in cup flow times but this is to be expected from Dacron-filled DAP due to the non-homogeneous quality of Dacron-filled formulations. Average flow time was 21.61 seconds at 10 tons ram pressure.

*Registered trademark of Food Machinery and Chemicals Corp.
+Registered trademark of E. I. duPont

ADHESIVES Forty-four formulations were prepared using Adiprene[+] 100, 167 and 213 resins in conjunction with polyol solutions of ferric acetyl acetonate (FAA) as a catalyst. The polyols used were 1,3-butanediol, 1,4-butanediol, 1,2,6-hexanetriol, 1,5-pentanediol, and 1,1,1-trimethylolpropane.

Most of the formulations contained polyol in excess of that present in the catalyst solution. At first, this solution contained 0.26 grams FAA per 10 grams of polyol. The normal formulation required 1.1 grams of this solution; hence, to 25 grams Adiprene resin was added 0.0281 gram FAA and 1.0719 grams polyol. The resulting adhesive formulations were evaluated by the following considerations in the order of importance: (1) elastomeric qualities of the film, (2) cure time, (3) pot life, (4) viscosity of adhesive mixture during duration of pot life, and (5) brushing characteristics of the adhesive. The first eleven formulations were the base for the remainder. All were quite viscous after degassing.

Variations were next prepared using excess polyol as a viscosity modifier. Most of these formulations required many days of cure and some did not cure at all. In an attempt to shorten cure time, twice the amount of catalyst solution was added to five formulations and five times the normal catalyst solution was added to four formulations.

The cure time did not decrease in any formulation where additional catalyst was used. The amount of polyol contained in the solution probably offset the effect of the additional catalyst.

A stronger concentration of FAA in the catalyst solution was prepared to control the effect of excess polyol. One gram of FAA was dissolved in 20 grams of polyol; thus, a normal formulation would contain 25 grams Adiprene resin, 0.0281 gram FAA, and 0.562 gram polyol; whereas the new formulation contains 25 grams resin, 0.0281 grams FAA and 1.0719 grams polyol. The evaluation of 44 formulations indicates that Adiprene 213 formulations are quite viscous and might be more difficult to apply by brushing; however, these formulations may be modified by adding Adiprene 167, or other formulations may be modified by the addition of Adiprene 213 to shorten cure time.

Also, it was noted that the use of excess polyol as a viscosity modifier must be held to narrow limits or the cure time will be excessive if any cure is obtained.

Those formulations which employ 1,1,1-trimethylolpropane (1,1,1-TMP) have produced films which are among the best obtained; yet, the adhesive solution is very viscous. Viscosity modification will require the use of other polyols than 1,1,1-TMP. Since it is a solid and adhesive, mixtures must be heated to get this compound into solution. The resulting adhesive solution is more viscous than one in which another polyol-FAA catalyst solution was used.

[+]Registered trademark of E. I. duPont

RADIOELEMENTS

Processes are being developed for separating and purifying radioelements and potential sources of supply are being evaluated.

SELECTIVE ADSORPTION OF TRACE ELEMENTS Work on the selective adsorption of trace elements on solid surfaces has been resumed on a limited basis. It was reported previously that, if a solution of actinium-227 in secular equilibrium with its decay products (AEM), is evaporated to dryness, one or more of the trace elements may be selectively desorbed and transferred, while the other elements remain adsorbed. As a rule, the elements are desorbed by solvents in which, in macro-amounts, they would be appreciably soluble, and remain inert in solvents in which they would be insoluble. Because the AEM solution is carrier-free, the concentrations of the principal radioelements (Ac^{227}, Th^{227}, and Ra^{223}) are far below those at which solubility products have any significance.

Results obtained thus far, while qualitatively consistent, have been erratic from a quantitative standpoint. This is probably due to lack of standardization in the conditions used, and the consistency can be expected to improve as experience is gained with regard to the significant variables.

In one group of experiments, an attempt was made to evaluate the effect of differences in the chemical nature of the surfaces used for adsorption: One milliliter of a 0.1 normal nitric acid solution of AEM, containing 0.15 microcurie of actinium-227, was transferred to each of three metal surfaces; stainless steel, gold-clad stainless steel, and platinum-clad stainless steel. One drop of concentrated ammonium hydroxide was added, and the solutions were evaporated to dryness.

The residual ammonium nitrate was evaporated from the stainless steel surface under an infrared lamp. However, because of the higher reflectivities of the gold and platinum surfaces, a high enough temperature could not be reached with an infrared lamp. The ammonium nitrate was driven off by heating the gold-clad and platinum-clad disks on a hot plate. Neither the temperature nor the heating time were closely controlled.

The sample disks were allowed to cool to room temperature. The residue on each disk was covered with one milliliter of recently boiled and cooled distilled water. The water was transferred to a second disk with the aid of a transfer pipet. The residue was covered twice more with one-milliliter portions of water and the water transferred to the second disk. The same operation was repeated with 0.005 normal nitric acid, and with 0.1 normal nitric acid. The solutions were evaporated to dryness, and the sample disks were ignited at red heat.

The sample disks were alpha-counted periodically, and the distribution of the radioelements was determined by the method of differential decay. The results are given in Table 1. The data show that, regardless of the surface metal, radium is more readily desorbed than actinium, and actinium is more readily desorbed than thorium. The higher retention of radium on the gold and platinum surfaces probably reflects the higher temperature at which the ammonium nitrate was driven off. Closer control of evaporation conditions will be required for evaluation of any differences due to the nature of the surface.

Somewhat better control was achieved with the following experiment: One milliliter of AEM solution was transferred to each of two sample disks; stainless steel and gold-clad stainless steel. No ammonium

hydroxide was added. The solutions were evaporated to dryness under an infrared lamp and the sample disks were allowed to cool to room temperature. Each residue was washed with a stream of distilled water from a wash bottle, until a total of 50 milliliters of water was collected. The leach solution was discarded. The sample disks were dried under an infrared lamp, but were not ignited. The samples were alpha-counted periodically, and the residual radioelements were determined by the method of differential decay. The results showed no significant differences between the adsorptive characteristics of the gold and stainless steel surfaces.

Another series of experiments was carried out in a similar manner; but, in this case various reagents were added either before or after the initial evaporation of the AEM solution. However, because the sample disks were not ignited before they were alpha-counted, there was considerable loss of activity in the chamber of the gas-flow proportional counter, and the results were ambiguous. This series will be repeated in the near future.

Table 1

FRACTIONATION OF Ac^{227}, Th^{227}, AND Ra^{223} FROM VARIOUS SURFACES

A - Stainless Steel

Leaching Agent	Per Cent Transferred Ac(III)	Th(IV)	Ra(II)
Distilled H_2O	28.7	5.1	91.4
0.005 N HNO_3	64.9	19.1	6.0
0.1 N HNO_3	3.8	64.4	0.5
Residual	1.1	9.7	0.3
Recovery	98.5	98.2	98.2

B - Gold-clad Stainless Steel

Distilled H_2O	8.4	5.8	57.0
0.005 HNO_3	79.7	10.7	39.4
0.1 N HNO_3	5.8	34.5	1.2
Residual	4.4	45.6	1.1
Recovery	98.3	96.6	98.7

C - Platinum-clad Stainless Steel

Distilled H_2O	16.8	1.2	76.0
0.005 N HNO_3	82.7	10.6	23.0
0.1 N HNO_3	1.4	24.5	0.4
Residual	1.7	58.4	0.7
Recovery	102.7	94.7	100.3

Although the results obtained thus far do not show quantitative separations of the actinium, thorium, and radium from each other, they have provided useful fundamental information with regard to the radiocolloidal behavior of these elements. The coprecipitation of radium-223 on cerous fluoride, for example, was reported last month. Confirmatory evidence was obtained when one milliliter of AEM solution containing one drop of 0.1 normal hydrofluoric acid was evaporated to dryness on a stainless steel disk. After the residue was cooled and washed with 50 milliliters of distilled water, the sample contained 43.1 per cent of the original radium-223. In the absence of hydrofluoric acid, no radium-223 was retained by the sample disk.

It is anticipated that similar experiments can be used to predict the coprecipitation of trace elements under various precipitating conditions.

HALF-LIFE OF RADIUM-223 Methods are being investigated for the preparation of carrier-free radiochemically pure radium-223. A carrier-free sample is desirable to permit its evaporation to the small volume required by the microcalorimeter.

A satisfactory separation of radium-223 has been made from an eight microcurie sample of actinium-227 in secular equilibrium with its decay products (AEM). The radium-223 yield was 82.7 per cent of theoretical, and analysis of the radium fraction showed that it contained approximately 0.01 per cent of the actinium-227. No thorium-227 was detected. The procedure is now ready to be tested at the millicurie level.

An analytical procedure has been developed for the separation and determination of traces of actinium-227 and thorium-227 in highly purified radium-223. It is now possible to determine these impurities after a few days of alpha counting time, instead of after one or two months. The procedure, which consists of three precipitations of lanthanum phosphate in the presence of barium holdback carrier, removes 97.8 per cent of the actinium and thorium is lost with each precipitation. The final precipitate, which has a counting yield of about 90 per cent of theoretical (before correction for self-absorption), contains 0.001 per cent of the radium-223. Working time is approximately one hour, and a preliminary result can be estimated six hours later, when the lead-211 has decayed to approximately 0.1 per cent of its initial value.

The purified radium-223 resulting from the eight microcurie separation is being used to make a preliminary determination of the half-life. The sample was purified further by the analytical procedure, and the radium-223 was recovered by barium sulfate precipitation. The barium sulfate has been mounted and is being alpha-counted daily.

The following procedure was used for the preparation of carrier-free radiochemically pure radium-223:

A small ion exchange column was prepared from a short length of soft glass tubing which had an inside diameter of 5.2 millimeters. The tip was drawn down to an opening of approximately two millimeters and plugged with glass wool. Dowex* 1 X8 resin (200-400 mesh) in the chloride form was added as a slurry in water to a settled height of approximately 40 millimeters. The resin column was washed with water, stirred with a fine glass rod and allowed to settle. The fine particles were removed, and the washing was repeated until the resin particles settled rapidly, leaving a clear supernate.

*Registered trademark of the Dow Chemical Company.

The resin was converted to the nitrate form by passing 50 per cent ammonium nitrate solution through the column until a sample of the effluent solution produced no turbidity with five per cent silver nitrate. The column was washed with approximately 10 milliliters of water.

Physical dimensions of the column were: inside diameter - 5.2 millimeters, cross-sectional area - 21.2 square millimeters, resin height - 34.0 millimeters, volume above resin - 1.88 cubic centimeters, flow rate with a full head of distilled water - 5.5 minutes per milliliter.

The source material was a 0.1 N nitric acid solution of actinium-227 in equilibrium with its decay products (AEM). One drop of concentrated ammonium hydroxide from a fresh stock bottle was added to one milliliter of AEM solution, and the mixture was allowed to stand at room temperature for approximately fifteen minutes. During the digestion period, the resin column was conditioned by the addition of one milliliter of freshly boiled and cooled distilled water to which was added one drop of fresh ammonium hydroxide. When the hydroxide appeared in the effluent (as indicated by a test with phenolphthalein), feeding of the AEM was begun. When all of the feed solution had been transferred to the column, the original vessel was washed twice with one milliliter of 1:20 ammonium hydroxide in boiled and cooled water. The washings were transferred to the column and the effluent was collected in a 10-milliliter volumetric flask.

The column was washed three times with one milliliter of boiled and cooled water, three times with one-milliliter portions of eight normal nitric acid, and three times with one-milliliter portions of boiled and cooled water.

Both of the distilled water fractions were transferred quantitatively to stainless steel disks and alpha-counted periodically. The ammonium hydroxide fraction was neutralized to phenolphthalein with one normal nitric acid, then diluted to ten milliliters with 0.1 normal nitric acid. The eight normal nitric acid fraction was diluted to ten milliliters with distilled water.

Aliquot portions of the original AEM solution and of the ammonium hydroxide and nitric acid fractions were taken to determine the yields of radium-223 and actinium-227. In addition, the ammonium hydroxide fraction was analyzed for actinium-227 and thorium-227 by the analytical procedures described below. The results of these analyses are shown in Table 2.

Table 2

DISTRIBUTION OF ACTINIUM-227, THORIUM-227, AND RADIUM-223[a]

Effluent Fraction	% Ac^{227}	% Th^{227}	% Ra^{223}
NH_4OH	0.01	-	82.7
H_2O	0.0015	0.007	0.4
8 N HNO_3	97.4	28.6	19.7
H_2O	2.6	1.2	0.5
Recovery	100.0	29.8	103.3

[a] Feed was eight microcuries of Ac^{227} in equilibrium with its decay products.

Although the primary objective of the present work is to prepare carrier-free radium-223, it is also desirable to recover the actinium-227 in a carrier-free form. Table 2 shows that this objective was also achieved. The loss of thorium-227 is not important, as this radionuclide grows into equilibrium with actinium-227 in six months. Although a higher yield of radium-223 would have been desirable, the data shown for the water fraction following the ammonium hydroxide elution indicate that a significantly higher yield would not have been obtained by further elution with ammonium hydroxide. The loss of radium-223 to the column is probably due to the presence of carbonate or sulfate in the source material, in the reagents, or in the column. If a trace of carbonate were present, it would also tend to improve the purity by forming radiocolloidal actinium and thorium carbonates, so such contamination would not be entirely undesirable.

Two principal problems remain; that of volume reduction by evaporation and sublimation of residual ammonium nitrate and the applicability of the procedure to quantities of actinium-227 approaching the milligram range. It is, therefore, planned that a test of the procedure will be made with approximately one millicurie of actinium-227 before the final run, involving 25 millicuries, is carried out.

In last month's report, an analytical procedure was described for the determination of traces of actinium and thorium in highly purified radium-223. In this procedure, the actinium and thorium are coprecipitated on ferric and cerous hydroxides in the presence of barium holdback carrier. The precipitate is centrifuged off and redissolved; barium holdback carrier is again added, and cerous fluoride is precipitated by the addition of hydrofluoric acid.

The hydroxide-fluoride separation recovered approximately 90 per cent of the actinium and thorium, while carrying 0.03 - 0.06 per cent of the radium. However, even this small amount of radium was sufficient to mask the alpha activity of the traces of actinium and thorium. It was necessary to alpha-count the cerous fluoride for three weeks before the method of differential decay could be applied to the estimation of the impurities.

Attempts to improve the radium separation by repeated precipitation and redissolution of the hydroxide resulted in progressive losses of actinium and thorium. Furthermore, it became apparent that thorium-227 was being lost to the walls of the glass centrifuge tube. A new approach seemed desirable.

If the pH of precipitation is carefully controlled, actinium and thorium may be separated from radium by coprecipitation on a rare earth phosphate. In the urinalysis procedure which is standard in this laboratory, cerous phosphate is precipitated at pH 4.5 to separate actinide elements from calcium and magnesium. Because of a tendency to oxidize to highly insoluble ceric phosphate on exposure to air, cerous phosphate does not lend itself well to repeated precipitation and redissolution. Lanthanum phosphate, however, does not suffer from this disadvantage and was adopted for use in the following separation procedure:

To the sample to be analyzed for actinium and thorium, add two milligrams of lanthanum carrier and ten milligrams of barium carrier. Add one drop of methyl orange indicator, two drops of 85 per cent phosphoric acid, and distilled water to a volume of about ten milliliters. Begin stirring.

Add concentrated ammonium hydroxide dropwise until the color is definitely yellow. Add one normal nitric acid dropwise until the color is just pink. Add one drop of one normal ammonium hydroxide (yellow color) and add 0.1 normal nitric acid dropwise until the color is just pink. (Note - the color change is not sharp,

and it is unwise to trust to memory with respect to the endpoint; hence, the repeated over-addition and back-titration of base and acid.)

Stir five minutes. Centrifuge five minutes, and discard the supernate (unless the radium-223 is to be recovered by precipitation of barium sulfate). To the precipitate add 10 milliliters of one normal nitric acid and begin stirring. Add 10 milligrams of barium carrier, one drop of 85 per cent phosphoric acid, and one drop of methyl orange indicator. Neutralize as before by the dropwise addition of concentrated ammonium hydroxide, one normal nitric acid, one normal ammonium hydroxide, and 0.1 normal nitric acid.

Stir five minutes. Centrifuge five minutes, and discard the supernate. Repeat the dissolution, additions, and precipitation as in the preceding paragraph.

Stir five minutes. Centrifuge five minutes, and discard the supernate. To the precipitate add 10 milliliters of 0.01 per cent dihydrogen ammonium phosphate solution. Stir vigorously five minutes. Centrifuge five minutes. Discard the supernate.

Mound the precipitate as a slurry in distilled water on a stainless disk previously prepared with a one-quarter inch retaining ring of collodion or Krylon plastic spray coating. Evaporate the slurry to dryness under an infrared lamp, and ignite the disk at red heat for 30 seconds. Allow six hours for the decay of lead-211, and alpha-count the sample periodically to determine the actinium-227 and thorium-227 by the method of differential decay.

The lanthanum phosphate procedure was evaluated with a standard AEM solution. The counting yield of actinium and thorium was between 88 and 93 per cent. Each of the supernatant solutions was found to contain only 0.5 - 0.7 per cent of the missing actinides, indicating that the loss in the main precipitate was probably attributable to self-absorption and geometry rather than to the chemistry of the separation.

The separation from radium-223 was evaluated with a portion of the purified radium-223 described earlier in this report. It was found that 97.8 per cent of the radium-223 was separated by each precipitation. After three precipitations in the presence of barium holdback carrier, the lanthanum phosphate contained 0.001 per cent of the original radium-223. Presumably, additional purification could be achieved, if necessary, by a fourth precipitation. The yield experiments indicate that there would be no significant loss of actinium or thorium.

Lead-211 appears to be coprecipitated quantitatively with lanthanum phosphate. Procedures could probably be developed to eliminate this nuclide, but the advantage to be gained by doing so seems negligible in comparison to the effort which would be required. By virtue of its 36.1-minute half-life, lead-211 decays to 0.1 per cent of its initial value within six hours, and a preliminary estimate of impurities can be made at that time, if necessary.

The analytical separation of traces of actinium and thorium isotopes from column-purified radium-223 resulted in a supernatant solution containing a doubly purified sample of radium-223. The radium-223 was coprecipitated with barium sulfate, and the precipitate was mounted for alpha-counting.

The decay of this sample has now been followed in the gas flow proportional alpha counter for approximately two weeks. Because of the high initial counting rate (approximately 2×10^6 counts per minute), the use of an accurately determined resolving time if of great importance. In an earlier report it was shown that the apparent half-life of a radioelement, as determined by counting, is highly dependent upon the value selected as the resolving time of the counter. It was assumed that the most accurate resolving time and half-life values were those which, when treated by the method of least squares, produced the smallest probable error.

The results of several least squares computations are shown in Table 3. Except for the case where τ, the resolving time, is 1.5×10^{-7} minute, all values of τ were arbitrarily selected. The value $\tau = 1.5 \times 10^{-7}$ minute was determined by the double source method, and represents the mean of 14 sets of observations with a probable error of 0.08×10^{-7} minute. The maximum observed τ was 1.745×10^{-7} minute at a counting rate of 845,000 counts per minute. The minimum τ was 1.291×10^{-7} minute at a counting rate of 237,000 counts per minute.

It will be noted that the half-life values reported in Table 3 range from substantially higher to substantially lower than the usually accepted value of 11.2 days, depending upon the value of the τ correction used. Inasmuch as the sample has been followed for less than two half-life periods, no conclusions can be drawn at this time regarding the most probable value of the half-life.

Table 3

HALF-LIFE OF RADIUM-223 AS A FUNCTION OF COUNTER RESOLVING TIME (τ)

(Spanning 16 days - 15 observations)

τ (Min. $\times 10^7$)	Half-Life (Days)	Probable error (Days)
1.5	11.6367	0.0262
1.7	11.3803	0.0207
1.9	11.1193	0.0150
2.0	10.9877	0.0128
2.2	10.7250	0.0108

IONIUM PROJECT A survey is being made of potential sources of thorium-230 (ionium) in the United States. Thus far, 49 samples have been received from 17 uranium mills or processing sites. In addition, several samples of "airport cake" from Mallinckrodt Chemical Works, retained from previous ionium processing, are available for comparison.

The procedures used to analyze the samples have been reported previously. A few minor changes have been made in the procedures to improve the speed of analysis; the changes will be incorporated in the report summarizing the completed survey.

Nineteen more samples have been analyzed, and the results are shown in Table 4. For two of the samples (10D and 22A) it was necessary to separate a precipitate from a clear supernate, and each fraction was analyzed separately. All of the solid samples analyzed this month have been too low in ionium content to be considered for processing. Five of the liquid samples have concentrations of 25 micrograms per gallon, or higher, but none are richer than the two samples (8D and 2A) which were previously reported to contain 74 and 120 micrograms per gallon, respectively.

Table 4

IONIUM IN URANIUM PROCESS SAMPLES

Identification	Ionium Concentration	Anal. Method	Ionium Potential of Source
6A (solid)	0.03 ppm	A	-
7A (solid)	0.03 ppm	B	-
8A (solid)	0.038 ppm	B	12.5 g./day
8B (solid)	0.017 ppm	A	-
8C (soln.)	44 μg./gal.	B	>100 g./day
10B (soln.)	28 μg./gal.	A	-
10C (soln.)	8.9 μg./gal.	A	-
10D-S (soln.)	11 μg./gal.	B	-
10D-R (solid)	0.01 ppm	A	-
10F (soln.)	nil	A	-
11A (solid)	0.02 ppm	A	-
13A (solid)	0.02 ppm	A	-
14A (solid)	0.01 ppm	A	-
15A (soln.)	28 μg./gal.	B	<10 g./day
15B (soln.)	9.1 μg./gal.	A	-
15C (soln.)	33 μg./gal.	A	-
20A (solid)	0.03 ppm	A	-
22A-S (soln.)	7.3 μg./gal.	B	-
22A-R (solid)	0.07 ppm	A	-
22C (soln.)	26 μg./gal.	A	-
23A (solid)	0.003 ppm	A	-

REACTOR FUELS AND MATERIALS DEVELOPMENT

Reactor fuels and materials development includes research, development includes research, development and design incident to the development of materials or techniques which lead to higher performance and/or lower costs for full scale power reactors.

Plutonium Alloy Research *Plutonium, which has a high neutron efficiency, is being considered for use in reactors of the fast breeder type. Mound Laboratory has been given the responsibility for acquiring data on some of the proposed fuel systems, and for maintaining technical cognizance of fuel cycle problems associated with the fast breeder reactors. Research has been initiated to determine the density, viscosity, thermal capacity, thermal conductivity and phase equilibria of plutonium and plutonium alloys proposed as fuels for these reactors.*

The viscosity of zinc was measured from 430 to 759°C at five different temperatures, and the viscosity of plutonium was measured up to 750°C. A different lot of plutonium metal was used in this work than in previous determinations.

The zinc determination was made in a sealed tantalum cup with 71.827 grams of 99.99 per cent pure zinc metal. The viscosity values are in agreement with previous data obtained at Mound Laboratory. The composite results of both determinations are presented in Table 5 and Figure 1. The straight line represents a least square fit for all 14 points reported. Literature values are included in Figure 1 for comparison. The Mound values represent a more extensive study of molten zinc than previously attempted by other investigators.

Table 5

VISCOSITY OF LIQUID ZINC

Run Number	Temperature (°C)	Density (g/cc)	Viscosity (centipoise)
1	823	6.206	1.72
2	759	6.260	1.77
1	716	6.306	1.96
2	660	6.356	2.07
1	601	6.414	2.28
2	553	6.456	2.54
1	496	6.516	3.07
2	480	6.531	3.12
1	446	6.562	3.66
1	432	6.574	3.68
2	430	6.580	3.37
1	428	6.578	3.72
1	421	6.582	3.71

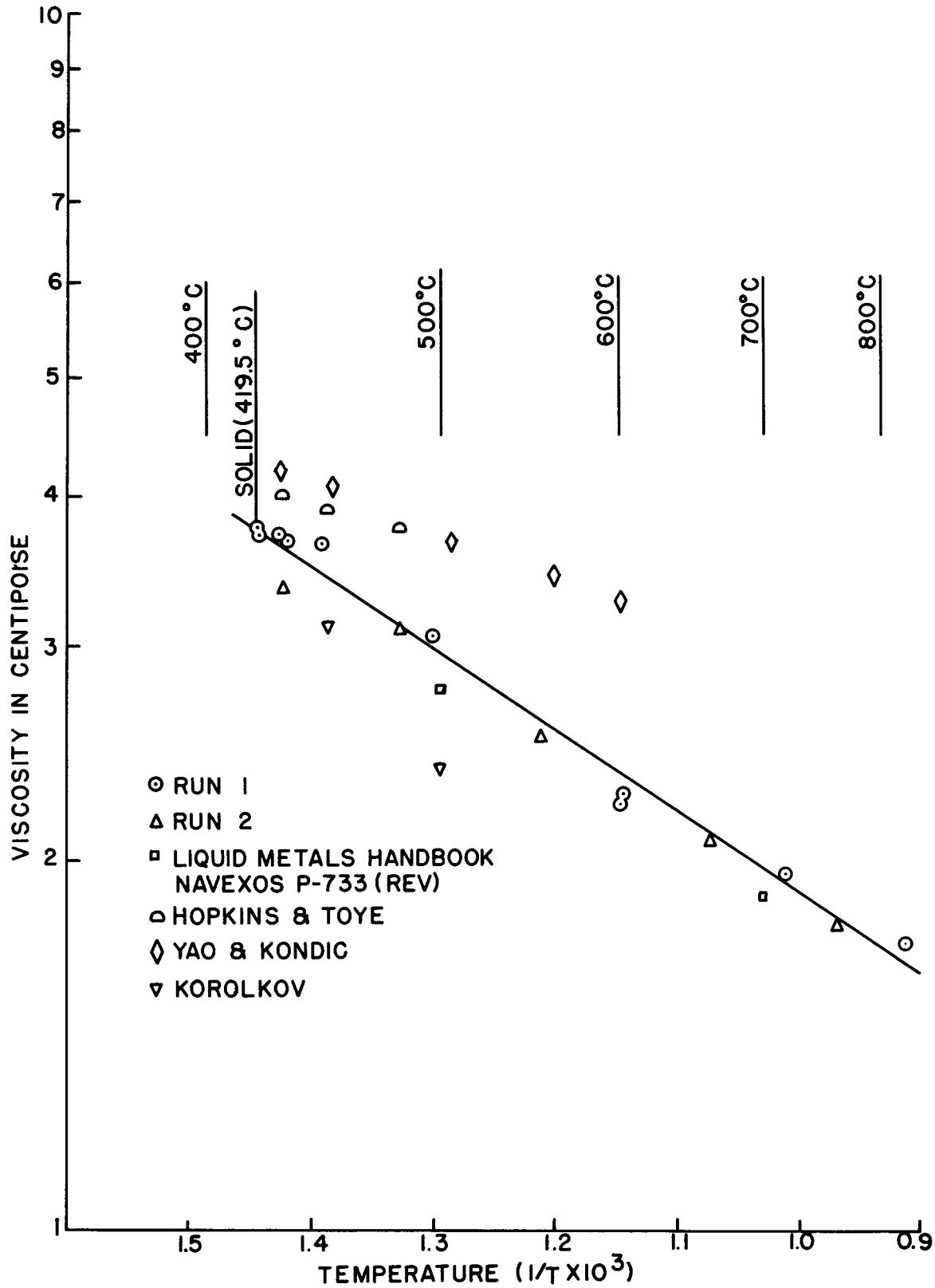

FIGURE 1 - VISCOSITY OF ZINC AS A FUNCTION OF ABSOLUTE TEMPERATURE

8785

The determinations of the viscosity of zinc in graphite-lined crucibles resulted in values that were 10 to 15 per cent lower than the values obtained with unlined crucibles. After this determination, the zinc casting was loose in the graphite liner and the liner walls showed no evidence of attack or wetting by the zinc. The lower viscosities may result from the non-wetting of the graphite by the molten zinc. Further investigation of the use of these liners is planned.

The investigation of the viscosity of the rare earth metals will be expanded to lanthanum and praseodymium for a comparison with the viscosity of cerium.

The viscosities obtained from the plutonium were lower than previous determinations. When the tantalum crucible was opened after the determination, examination revealed that the plutonium rod in the viscosimeter cup was badly oxidized and showed numerous voids in the top one-half of its length. The data obtained are unsatisfactory. Since such oxidation has not been observed previously, this determination will be repeated with a different lot of plutonium metal.

The determination of the density of liquid cerium has been initiated. Ten pycnometers, whose internal volumes were approximately 0.5 cubic centimeters, have been calibrated to within 0.01 per cent. The measurement of the density of a liquid plutonium-iron alloy has been scheduled to follow the cerium.

Metallographic examinations continued on the plutonium-copper binary system. All of the samples with composition above 55 atom per cent copper have been polished, etched, and photographed. Microhardness measurements of the different phases were made. Table 6 gives the compositions of the alloys, and the microhardnesses of the phases present. The only departure from the diagram determined by differential thermal analysis was in an 80.4 atom per cent alloy which was not taken from a DTA tube. This sample showed an extra phase present which cannot be rationalized by the phase diagram. This phase did not appear in samples taken from DTA samples, presumably because of the difference in cooling rates of the two methods. The DTA tubes are cut open after they have cooled at a rate of one to three °C per minute, whereas the alloy button, prepared in a magnesia crucible, was cooled considerably faster, probably at least 20 to 30°C per minute through the solidification range. The 80.4 atom per cent alloy shown in Table 6 was prepared by the alloy button technique, which may explain the high hardness values for the phases of this sample. Until the existence of this new phase is definitely established, the phase is listed in the table as "PuCu or unknown" phase. Further investigations are planned to resolve the problem.

A nickel block has been turned by the machine shop to fit into the high temperature calorimeter. This block will supply a constant temperature bath around the calorimeter forms and will replace the silver block originally used as the isothermal bath.

The calorimeter bridges wound on the beryllium oxide forms have been balanced and are now being leached in solutions of acetone and benzene, preparatory to being fired.

Several blocks of diatomaceous earth have been prepared to be used as thermal gradient media for the calorimeters. Two of these have been fired. Entrapped air bubbles, which had apparently been formed in the mixing of the diatomaceous earth with water, were found in the first series of blocks. These blocks were discarded as they would not have provided a smooth temperature gradient. Of the two which were fired, the second was found to have good thermal characteristics, high electrical resistivity and was easily machined to the proper shape.

Table 6

PHASE IDENTIFICATION IN Cu-Pu SYSTEM

Composition	Condition of Sample	Phase Identified	VHN
		Pu_4Cu_{17}	509-540
80.4	Annealed	$PuCu_4$	585-642
		$PuCu_4$ or unknown	560-599
80	Annealed	$Pu_4Cu_{1.7}$	447-480
		$PuCu_4$	423-480
78	As Cast	Pu_4Cu_{17}	460-501
		$PuCu_4$ or unknown	487-532
78	Annealed	$PuCu_4$ or unknown	468-540
		$PuCu_2$	357
76	Annealed	$PuCu_4$ or unknown	425-464
		$PuCu_2$	353
67	As Cast	$PuCu_2$	387-397
		$PuCu_4$ or unknown	308-404
67	Annealed	$PuCu_2$	363-383
		$PuCu_4$ or unknown	429-536
55	As Cast	$PuCu_2$	380-401
		α Pu	108-194
55	Annealed	$PuCu_2$	333-413
		α Pu	117-181

Plutonium-Bearing Glass Fibers *At the request of the Reactor Fuels and Materials Development bracnh of the AEC, a cooperative experiment in the fabrication of plutonium-bearing glass fibers is being carried out by Rensselaer Polytechnic Institute, Owens-Corning Fiberglas Corporation and Mound Laboratory.*

An experimental sodium silicate glass containing aluminum oxide, calcium oxide, potassium oxide, and 10 weight per cent plutonium oxide was completely homogenized. This glass was drawn into fibers by the monofilament process without difficulty. Leach tests of the glass fibers in water and in 0.1 normal hydrochloric acid are now in progress. Preliminary results indicate a significant increase in stability compared to the glass composition previously tested. The dissolution of the glass by 0.1 normal hydrochloric acid appears to be greater than by water alone, although after 43 hours both rates of attack appear to be very low.