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Mound Laboratory - Monsanto

Central File No. 57-5-42

Contract Number AT-33-1-GEN-53

MOUND LABORATORY

Operated By

MONSANTO CHEMICAL COMPANY

MIAMISBURG, OHIO

MONTHLY TECHNICAL ACTIVITIES REPORT

THROUGH MAY 15, 1957

By

J. F. Eichelberger and D. L. Scott

[redacted]

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Date: May 16, 1957

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PRODUCTION PROGRAM

These projects are concerned with the production of alpha and neutron sources, and with the disposal of radioactive wastes. Work is being done to improve present methods and processes.

Nine polonium alpha sources, ten polonium-beryllium sources, and eighteen plutonium-beryllium sources were shipped during April. Five polonium-beryllium sources and one plutonium-beryllium source were for other AEC sites.

Twenty-five tanks of influent water were processed during the period. Eleven of the tanks required no pretreatment, while four tanks required extensive treatment with barium chloride and sodium sulfide. An extraordinary large amount of influent water was received in one day during the early part of May 1957 which required that the "B" clariflocculator be used earlier than expected.

Data for the Waste Disposal Operations are given in Table I.

TABLE I

Discharge Volume	563,400	gallons
Total Alpha Activity	6.9	millicuries
Total Beta Activity	19.8	millicuries
Activity Density		
Alpha	3.6	cts/min/ml
Beta	10.3	cts/min/ml

NUCLEAR RESEARCH AND DEVELOPMENT

CHEMISTRY AND METALLURGY

Chemical and metallurgical studies applicable to the weapons program are being made. These studies are directed towards the production of several radiochemically pure elements and of a chemically pure plastic material.

Ionium Project

The ionium project is directed toward the development of a process for the extraction of ionium from partially processed raffinates from the Mallinckrodt Uranium Refinery.

Processing - The solution containing 120 - 150 grams of thorium-230 was received from Mallinckrodt Chemical Works. The information received with the solution indicates that it also contains about 16 kilograms of uranium in one-molar sodium sulfate solution. Some data on extraction from a sulfate solution have also been received from Mallinckrodt Chemical Works. These data are being studied, and the best method for accomplishing the additional operations is being considered.

Present plans indicate that one month will be required for complete processing of this material. However, since a sulfated solution has never been processed in the mixer-settler units, the time required may vary from one week less to one month more.

Mixer Settler Development - Work on the small, glass mixer-settler units has been temporarily stopped in favor of higher priority projects.

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Protactinium Project

The Albuquerque Operations Office has authorized the production of a gram of protactinium-231 from the unprocessed Sperry-Filter-Press residues from Mallinckrodt which remain after the isolation of the first gram.

Production Facilities - "Hot" processing started on schedule on April 17. However, the project is now about two weeks behind schedule because, for the most part, of mechanical and chemical failures normally encountered in the start-up of a new facility. The mechanical failures should be corrected by May 15 at which time the facility will be operated on a two shift basis.

The chief chemical problem was the decrease in concentration of the DIBC in the solvent mixture. This problem had not been noticed before since the solvent had not been cycled sufficiently in any test. An analytical method for determining the per cent DIBC in the DIBC-Amsco mixture was developed so that the additional DIBC required could be estimated. The analytical method is reported later in this section. The decrease in DIBC concentration is shown in Table I.

TABLE I

Decrease in DIBC Concentration

On-Stream Time Hours	DIBC Concentration Per Cent
0	44
2.2	46
4.2	43
6.2	42
8.7	40
10.7	40
14.9	36
17.3	34
20.8	26

As a result of this decrease, the aqueous raffinates from the initial processing will be recycled. Although the DIBC concentration decreased, the iron was nearly completely removed in the first pass. Iron will therefore not be present to interfere with the raffinate recycle. A total of eleven drums or approximately 5500 pounds of Sperry-Cake have been digested and processed in the columns.

Protactinium Purification - The protactinium concentrate recovered from operations during Fiscal Year 1956 is distributed among several vessels in both aqueous and organic media. Except for various residues and precipitates containing small amounts of protactinium, nearly all of the protactinium originally in solution has hydrolyzed and precipitated. This situation makes it extremely difficult to obtain an accurate estimate of the amount of protactinium in each container, but it was previously estimated that the total protactinium in the concentrates was about 700 milligrams.

This hydrolytic precipitation of protactinium has, however, provided a fortuitous separation from large quantities of iron which have persisted throughout the concentration from the raffinate residue. The supernatant liquor is easily decanted because the precipitate tends to stick to the walls of the polyethylene vessels in which the solutions were stored.

Although the protactinium-bearing residue responds only slowly to digestion with concentrated sulfuric acid, heat cannot be applied because of the low softening point of polyethylene. Repeated attack with sulfuric acid ultimately results in the complete removal of protactinium from the container. Once in solution, the protactinium is readily extractable into diisobutyl carbinol.

The protactinium in the supernatant liquors from the hydrolytic precipitates can be recovered by phosphate precipitation. The freshly precipitated protactinium dissolves easily in sulfuric acid, and can be extracted into diisobutyl carbinol

Work on the consolidation of the protactinium in a homogeneous solution has been started. An estimated 50 milligrams of protactinium is now in solution in diisobutyl carbinol, and about one month will be required to complete the operation. Waste solutions from this operation are being accumulated and will be combined with solutions being obtained in the current protactinium recovery process. When this phase of the work has been completed, purification should proceed rapidly on the basis of experiments described in last month's report

Approximately 15 milligrams of purified protactinium in solution was shipped to Los Alamos Scientific Laboratory for fast neutron cross-section studies. A spectrographic analysis of a sample of this solution will be made as soon as the equipment for firing radioactive samples is available

Analytical - After it became evident that a method was needed to determine the per cent DIBC in Amsco, a study was made of the physical properties of the two components. The densities and boiling points of DIBC and Amsco solvent were too similar to be the basis for a reliable method. Data on the viscosities were not available. Attention was therefore turned to the chemical properties. Esterifying the DIBC with acetic anhydride gave erratic results. It was noted that upon shaking a DIBC-Amsco solution with concentrated sulfuric acid that a lower aqueous layer of sulfuric acid, an intermediate layer of DIBC, and an upper layer of Amsco formed. The two upper layers were not very stable. It was decided to try adding a component having a high distribution coefficient to the organic. When an organic sample was shaken with a sulfuric-hydrochloric acid solution containing ferric ion, three very stable layers were formed. The layers could be re-mixed and they immediately separated by gravity. Besides being stable, the upper layer was clear amber and the middle layer was a very dark brown so that the interface was very easily seen

Reagent grade chemicals are used throughout. The solution containing iron is prepared as follows:

> To 250 ml of concentrated HCl, finely ground $FeCl_3 \cdot 6H_2O$ is added with constant stirring until the volume increases to 375 ml. The dissolution is endothermic and undissolved salt remains in the solution. The solution is stirred constantly and 125 ml of cold, concentrated H_2SO_4 is added with a burette with the tip submerged in the solution. The resulting solution is about 9 *N* in H_2SO_4 and 6 *N* in HCl, and contains about 80 mg of iron per milliliter.
>
> The determination of per cent DIBC is made as follows: Ten ml of the sample is pipetted into a 100 ml volumetric flask and is diluted to the mark with 100 per cent Amsco, a highly refined kerosene. Fifty ml of this solution is pipetted into a 125 ml separatory funnel. Fifteen ml of the iron solution is added to the funnel and the funnel is shaken for one minute, relieving any pressure by periodically venting through the stopcock. After allowing to settle for one or two minutes, the solution is transferred to a clean, dry 50 ml burette, and the aqueous layer is drained to

permit all of the organic phases to be retained in the burette. The separatory funnel is rinsed with H_2O and shaken, and the contents are drained into the burette. The clear Amsco layer is drained nearly to the stopcock and H_2O is added to raise the layer to about ½-inch above the zero mark on the upper end of the burette. A rubber stopper is placed in the burette and the burette is inverted several times until the walls have been rinsed with H_2O. The burette is drained until the Amsco is about ⅛-inch above the zero mark and allowed to stand for 10 minutes. The solution is drained down to the zero mark and the lower Amsco meniscus reading is recorded. The uncorrected per cent DIBC is then given by the following equation:

$$\text{Per cent DIBC by volume} = \frac{100 - 2R \times 100}{10}$$

where R is the burette reading

This method gives the true value at 60 per cent DIBC. Values from zero to 60 per cent read low, and values from 60 to 100 per cent read high Pure Amsco reads zero per cent DIBC and pure DIBC reads 110 per cent DIBC. Standards were prepared and a plot was made of per cent DIBC by test versus per cent DIBC by standard. The validity of the plot was checked by spiking samples after test to bring them to 60 per cent and re-testing. This spiking procedure indicated that the corrected value was within ± 2 units of the correct value.

Errata

In CF-57-4-32, "Monthly Technical Activities Report Through April 15, 1957", page 7, third paragraph, line 9, should read, "yielded a small precipitate and a supernate which was clear except for a few small, floating particles ....."

Plutonium Urinalysis

Present methods for determining the amount of plutonium in human urine give unsatisfactory results. A study directed towards adaptation of the present successful procedure for actinium-thorium urinalysis to the recovery and determination of plutonium is being made.

There is no further progress to report on this project at this time.

Plastics Project

This project is immediately directed toward developing a satisfactory process for the blending of diallyl phthalate powder and filler into a plastic whose physical and chemical properties are suitable for the molding operations at Mound Laboratory. When this process has been satisfactorily developed, a search for materials and processes which will provide superior physical and chemical properties will be begun.

Plastics Formulation - Five formulations (B38 through B42) were made in the sigma-blade mixer. All but B42 have been milled and are being evaluated. The compositions for formulations B36 through B42 are given in Table I.

TABLE I

Diallyl Phthalate Formulations

Dapon plus Filler, 25 pounds; Monomer, 5 parts

Run No.	Filler Used	Parts Per 100 Parts of Dapon 35						Residual Acetone Per Cent
		Filler	Acetone	Catalyst a/b	Zinc Stearate	Pigment c/d	TiO_2 e/f	
B36	Cellulo 200 Hydrite PD 121	40 60	250	2.0 "	2.0	" "	" "	13.5
B37	Cellulo 200 Hydrite PD 121	40 60	250	2.0 "	2.0	" "	" "	11.3
B38	Cellulo 200 Hydrite PD 121	40 60	225	1.8 0.2	2.0	" "	" "	10.2
B39	Cellulo 200 Hydrite PD 121	40 60	200	2.0 "	1.5	2.0 "	" "	12.6
B40	Cellulo 200 Hydrite PD 121	40 60	200	1.8 0.4	1.5	2.0 "	0.5 "	10.8
B41	Cellulo 200 Hydrite PD 121	40 60	200	1.75 0.5	1.5	2.25	0.25 "	10.8
B42	Cellulo 200 Hydrite PD 121	40 60	200	1.75 0.5	1.5	2.0 0.2	0.3 "	5.6[g]

a t-butyl perbenzoate
b di-t-butyl peroxide
c Scarlet 25AD, IR 6440
d Madder Lake R, IR 6050
e Rutile
f Anatase
g partially polymerized

Filtration of the DAP varnish was again attempted with run B38. A Fiberglass filter-cloth was clamped between two nickel-plated brass rings, the lower of which was silver-soldered to the stainless steel funnel of the sigma-blade mixer. The cloth was supported by a nickel-plated, open-mesh screen. The initial filtration rate was fairly good, but the rate became very slow before one-half of the varnish had been added. The remaining varnish was therefore added without filtration.

Stirring overnight resulted in a reasonably fluid mass with the same decrease in input power observed during run B37. There was a greater tendency for the plastic mass to ride on the sigma-blades in run B38 (225 parts of acetone) than in run B37 (250 parts of acetone), although fairly good kneading was observed during most of run B38.

Formulation B39 was run using 200 parts of acetone and was the first sigma-blade mixer run in which pigment was added. The pigment was added as a suspension in acetone in the same manner as the zinc stearate is normally added. Visual observation indicated rapid dispersion; no variation in color could be detected after a few minutes of stirring.

The power reduction on stirring overnight and the kneading action obtained indicated that a significant amount of defelting of the asbestos had been obtained. Kneading action was much poorer during the latter part of the run than for either B37 or B38.

A residual acetone content of 10 to 11 per cent gives a product which handles well on the mill rolls. Lower acetone contents could be used on the mill, but are probably not practical in the sigma-blade mixer.

Reasonably good correlation between input power to the sigma-blade mixer and residual acetone content has been obtained. Much better correlation would be expected if good kneading action could be maintained up to the end point. It is therefore felt that more accurate control, through power recording, will be obtained with production-size equipment than can be obtained with the present laboratory equipment.

In run B42 the residual acetone content was reduced to too low a value, and the temperature rise due to mechanical work became sufficiently great to initiate polymerization. The power required rose very rapidly, and the mixture became so stiff that the drive shaft was sheared. This accident will delay pilot plant production of DAP molding compound. Run B42 was the second run of the first attempt to produce 10 pounds of molding compound per day on a continuing basis.

Formulations B38 through B41 were milled on the differential rolls in approximately four-pound lots to lower values of input power than most previous runs. The net power consumed by the plastic at the time of cut-off and the type of guides used are given in several of the tables.

The input power to the No. 1 mill, which is fitted with plated guides, is essentially constant at 0.50 kilowatt when unloaded. However, the power input to the unloaded No. 2 mill, which is fitted with Nylon guides, has varied from 0.50 to 0.60 kilowatt. This variation is presumably caused by a variable friction between the Nylon guides and the rolls. Completion of the milling is now judged by the net power consumed by the plastic (total input power minus the unloaded input power) rather than the input power itself as was used previously.

Additional data on the variation of flow time with milling conditions, and with the age after grinding, are given in Tables II, III, IV, V, and VI.

TABLE II

Variation of Flow Time Between Samples Milled to Definite Power Inputs
B35-2, B35-3, B35-4, B35-5 and B35-6

With plated guides and 0.176 inch roll spacing

Hours After Grinding	Flow Times at 5 Tons, and Deviations, in Seconds									
	B35-2 to 1.32 KW	B35-3 to 1.33 KW	B35-4 to 1.32 KW	B35-5 to 1.32 KW	Maximum Deviation Plus	Maximum Deviation Minus	Maximum Spread	Average[a] A_{25}	B35-6 to 1.23 KW A_6	A25-A_6
1	3.78	3.17	3.61	3.77	0.20	0.41	0.61	3.58	3.68	−0.10
2	3.36	3.72	3.46	4.12	0.45	0.31	0.76	3.67	5.59	−1.92
20	6.35	6.52	7.04	5.95	0.57	0.52	1.09	6.47	4.36	0.59
48	7.85	7.20	6.86	7.64	0.46	0.53	0.97	7.39	6.54	0.85
120	10.38	9.47	9.40	9.90	0.59	0.39	0.98	9.79	8.40	1.39
188	9.81	9.23	9.75	9.42	0.26	0.32	0.58	9.55	9.53	0.02
288	11.59	10.25	10.87	11.10	0.64	0.70	1.34	10.95	10.55	0.45
307	-	-	-	-	-	-	-	-	10.43[b]	-
504	12.45	12.32	12.13	12.39	0.13	0.19	0.32	12.32	12.50	−0.18

a Average of B35-2, B35-3, B35-4 and B35-5

b Measured before being preformed for production molding.

c Estimated ultimate flow in seconds at 5 tons; B35-2 through B35-5, = 18; B35-6 = 16.

TABLE III

Variation of Flow Time Between Samples Milled to Definite Power Inputs

With Change in Roll Spacing During Milling[a]
B36-1, B36-2 and B36-3; B36-4, B36-5 and B36-6

Hours After Grinding	Flow Times at 5 Tons, and Deviations, in Seconds									
	No. 1 Mill with Plated Guides					No. 2 Mill with Nylon Guides				
	B36-1 to 1.21 then to 1.44 KW	B36-2 to 1.21 then to 1.41 KW	B36-3 to 1.22 then to 1.51 KW	Maximum Spread[b] Δ_{13}	Average[c] A_{13}	Average[d] A_{46}	B36-4 to 1.18 then to 1.43 KW	B36-5 to 1.14 then to 1.34 KW	B36-6 to 1.14 then to 1.33 KW	Maximum Spread Δ_{46}
1	3.66	3.91	4.03	0.37	3.87	3.10	3.30	2.80	3.20	0.50
24	4.57	3.98	4.71	0.73	4.42	4.42	4.45	4.21	4.61	0.40
48	4.76	5.69	5.48	0.93	5.31	5.36	5.38	5.41	5.30	0.11
72	6.25	5.70	6.58	0.88	6.18	6.07	6.01	5.88	6.31	0.43
144	7.90	7.70	8.00	0.30	7.87	7.36	7.63	7.24	7.20	0.43
336	8.08	8.91	8.85	0.83	8.61	9.00	9.04	8.91	9.04	0.11
504	8.93	9.63	10.13	1.20	9.56	9.57	9.50	9.32	9.89	0.49

a Milled at 0.176 inch roll spacing to first power given then rolls closed and milling continued until second power value was reached.

b The maximum spread between all six materials varied between 0.73 and 1.23 seconds, average 0.92 seconds.

c The subscript 13 indicates B36-1, B36-2 and B36-3.

d The subscript 46 indicates B36-4, B36-5 and B36-6.

TABLE IV

Variation of Flow Time Between Samples Milled to Definite Power Inputs

With change in Roll Spacing During Milling[a]

B37-1 through B37-6[b]

Hours After Grinding	Flow Times at 5 Tons and Deviations, in Seconds									
	B37-1C[c] to 1.25 then to 1.55 KW	B37-2C to 1.30 then to 1.50 KW	B37-3C to 1.25 then to 1.55 KW	B37-4C to 1.25 then to 1.52 KW	B37-5C to 1.25 then to 1.55 KW	B37-6C to 1.25 then to 1.55 KW	Average A 16	Maximum Deviation Plus	Maximum Deviation Minus	Maximum Spread
72	5.30	6.27	4.91	5.46	5.32	5.35	5.44	0.83	0.53	1.36
96	6.18	6.54	6.23	5.91	5.45	5.11	5.90	0.64	0.79	1.43
168	7.69	6.93	7.11	6.96	6.67	6.90	7.04	0.65	0.37	1.02
288	8.76	8.19	8.12	8.36	7.99	8.42	8.31	0.45	0.32	0.77
336	8.42	7.97	8.31	8.24	8.36	7.68	8.16	0.26	0.48	0.74

a Milled at 0.176 inch roll spacing to the first power given, then rolls closed to 0.080 inch and milling continued until the second power value was reached.

b Milled in No. 2 mill using Nylon guides.

c Symbol C indicates that material finer than 100 mesh has been removed.

TABLE V

Variation of Flow Time Between Samples Milled to Definite Power Inputs[a]

B38-1C through B38-6C

Hours After Grinding	Flow Times at 5 Tons, and Deviations, in Seconds											
	B38-1C to 1.25 KW	B38-4C to 1.25 KW	B38-5C to 1.26 KW	B38-6C to 1.23 KW	Maximum Deviation Plus	Maximum Deviation Minus	Maximum Spread	Average[b] A_1	B38-2C to 1.35 KW A_2	A_1-A_2	B38-3C to 1.11 KW A_3	A_1-A_3
24	4.74	4.43	4.50	4.97	0.31	0.23	0.54	4.66	4.65	0.01	4.19	0.47
144	7.28	7.11	7.04	7.37	0.17	0.16	0.33	7.20	7.18	0.02	7.30	-0.10
336	9.38	9.65	9.15	9.34	0.27	0.23	0.50	9.38	9.24	0.14	9.91	-0.53

a Milled on No. 2 mill with Nylon guide, 0.176 inch roll spacing.

b Average of B38-1C, B38-4C, B38-5C, and B38-6C.

c Material finer than 100 mesh removed from these samples.

TABLE VI

Variation of Flow Time Between Samples Milled to Definite Power Inputs[a]

B39-1C and B39-3C, B39-2C, B39-4C and B39-6C

Hours After Grinding	Flow Times at 5 Tons, and Deviations, in Seconds								
	B39-1C to 1.18 KW	B39-3C to 1.19 KW	Average A1-3	Average A 46	B39-4C to 1.13 KW	B39-5C to 1.13 KW	B39-6C to 1.14 KW	Maximum Spread Δ_{46}	B39-2C to 2.7 KW[b]
72	5.85	5.64	5.75	7.32	6.96	7.55	7.46	0.59	6.86
144	7.83	8.82	8.33	9.20	9.12	9.50	8.99	0.43	8.22
240	8.90	9.28	9.09	9.43	9.20	9.55	9.54	0.35	9.18
336	10.62	10.87	10.75	10.29	10.58	10.01	10.28	0.57	9.84

a Mill No. 2, Nylon guides, 0.176 inch roll spacing.

b By accident mill roll spacing was 0.010 inch at one end and 0.176 inch at the other.

c Material finer than 100 mesh removed from these samples.

Interpretation of these data is difficult, but an attempt at a partial analysis will be made, taking into consideration all available information

It would appear that the rate of change in flow with time is a complex function of the ultimate flow. As reported last month, relatively stiff material gives a straight line with some scatter of the points when the flow time in seconds is plotted against the logarithm of the age, but tends to deviate from linearity at short and at very long times For softer materials, a plot of the logarithm of the flow time in seconds against the logarithm of the age appears to give a better straight line except at short times.

Stiff materials tend to reach an approximately constant flow which may be much lower than the ultimate flow; hence, plots on linear coordinates are of doubtful value. Softer materials also tend to reach a pseudo-equilibrium, but the difference between the pseudo-equilibrium and the ultimate flow probably becomes progressively less as the ultimate flow is decreased, and plots on linear coordinates may be useful for softer material It may be that a material of lower ultimate flow could give a longer closure time in the cup-flow test than a material of higher ultimate flow at some intermediate age. Possible examples of this are B38-2C and B38-3C in Table V Some other data in the tables which appear unreasonable can probably be reconciled if the above assumptions are correct.

An examination of Tables II, III, IV, V, and VI indicates that adequate control of mill time can probably be obtained by the use of the recording wattmeter. The rate of power increase near the end point may vary over a rather wide range; hence, the time required to mill to a definite ultimate flow may also vary widely. There is a rough correlation between the change in the surface temperature of the plastic and the change in input power, and measurement of the surface temperature of the plastic has proved helpful.

The folding of the plastic into the rolls by the guides cause the input power to vary in an approximately periodic manner so a smooth power curve is not obtained It is therefore necessary to determine, by examination of a length of the curve, the power level above and below which equal areas are drawn in order to determine the average power The average power, so determined, has been found to be considerably more consistent than the peak power. However, the peak power reached from time to time is helpful in determining when the end point is near.

The problem of determining the ultimate flow or some other satisfactory property in a reasonably short time after grinding has yet to be solved. Since the finer fractions increase in flow time and approach an approximately constant value more rapidly than coarse fractions, early measurements on a fine fraction may be one possible solution. Present screening facilities are entirely inadequate to permit an extended study of this possibility. A gyro-centric screen has been placed on order. This will make routine removal of fine material practical for pilot plant production, and will make available much larger quantities for study.

The removal of fines from all plastic intended for production molding should minimize certain present difficulties.

Further data on the variation of flow time with particle size and with age after grinding are given in Tables VII, VIII, IX and X

TABLE VII

Variation of Flow Time with Particle Size and with Age After Grinding

B35-1 Milled with 0.176 inch Roll Spacing to 1.52 Kilowatt Net Input Power

Sample No. B35-	1	S1[b]	S2	S3	S4	S5	S9/11
Mesh Range	a	-8 +16	-16 +20	-20 +40	-40 +60	-60+80	-140
Av. Particle Diam in microns	a	1780	1015	630	335	213	70
Per Cent of Total	100	24.77	31.82	26.43	7.20	3.68	3.31
Hours After Grinding	Flow Time[c]	Flow Time	Flow Time	Flow Time	Flow Time	Flow Time	Flow Time
1	4.65	-	-	-	-	-	-
6	5.60	4.10	4.69	6.74	-	-	-
72	8.75	7.50	8.40	9.49	11.50	-	-
168	9.09	7.96	9.39	10.61	12.20	13.17	13.05
288	10.41	-	-	-	-	-	-
336	-	9.04	10.22	11.74	-	-	-
408	10.92	9.41	10.18	12.78	-	-	-
576	11.71	10.36	11.16	-	-	-	-
768	11.49	-	11.15	-	-	-	-

a Ground in Cumberland Grinder using 3/32 inch screen

b S Fractions are B35-1 screened with Rotatap.

c Flow times in seconds at 5 Tons pressure.

d Estimated ultimate flow 18 to 20 seconds.

TABLE VIII

Variation of Flow Time with Particle Size and with Age after Grinding

B35-2/5S Milled with 0.176 inch Roll Spacing to 1.32 KW Net Input Power

Sample B35-2/5	Per Cent of Total	Mesh Size Range[a]	Flow Times, at 5 Tons, in Seconds At 24 hr	At 48 hr	At 116 hr	At 164 hr	At 284 hr	At 336 hr	At 552 hr	At 672 hr
-S1	24.96	-8 +16	4.53	5.00	7.00	8.12	8.66	9.38	10.11	10.28
-S2	29.08	-16 +20	5.41	6.14	8.03	8.87	9.34	10.31	11.30	11.63
-S3	26.28	-20 +40	6.02	7.62	9.96	10.26	12.36	11.80	12.74	12.97
-S4	7.73	-40 +60	10.30	12.18	12.00	11.74	13.12	13.10	-	13.77
-S5	4.14	-60 +80	10.95	12.95	14.26	-	-	-	-	-
-S6	1.73	-80 +100	13.34	-	-	-	-	-	-	-
-S7	1.02	-100 +120	12.36	-	-	-	-	-	-	-
-S8	0.97	-120 +140	11.99	-	-	-	-	-	-	-
-S9/10	1.05	-140 +200	-	12.60	-	-	-	-	-	-
-S11	3.06	-200	12.75	-	12.85	-	-	-	-	-

a Ground in Cumberland grinder using 3/32-inch screen, classified by use of Rotatap.

TABLE IX

Variation of Flow Time with Particle Size and with Age After Grinding Blend of B36-1 through B36-6[a]

Sample B36-1/6	Per Cent of Total	Mesh Size Range[b]	Flow Times at 5 Tons, in Seconds at 20 hr	at 24 hr	at 48 hr	at 72 hr	at 168 hr	at 384 hr	at 504 hr
-S1	25.06	-8 +16	2.90	3.51	3.54	4.31	6.19	7.42	8.28
-S2	30.45	-16 +20	3.85	4.21	4.25	5.38	6.91	8.09	9.12
-S3	26.58	-20 +40	5.75	5.53	5.52	6.74	7.91	9.11	9.55
-S4	7.58	-40 +60	8.56	7.34	8.36	8.83	9.56	10.04	11.78
-S5	3.76	-60 +80	-	9.07	-	10.37	10.64	10.01	-
-S6	1.54	-80 +100	-	-	-	10.25	-	-	-
-S7	0.79	-100 +120	-	-	-	-	-	-	-
-S8	0.82	-120 +140	-	-	-	-	-	-	11.73
-S9	0.87	-140 +200	-	-	-	-	-	-	-
-S10	2.55	-200	-	8.74	-	-	10.27	-	11.90

a For conditions of milling see Table III.

b Ground in Cumberland Grinder 3/32-inch screen, classified by use of Rotatap.

TABLE X

Variation of Flow Time with Particle Size and with Age After Grinding Blend of B41-1 through B41-6[a]

Sample B41-1/6	Per Cent of Total	Average Diameter Microns	Mesh Size Range[b]	Flow Times at 5 Tons, in Seconds at 73 hr	at 120 hr	at 240 hr
-S1	1.31	1780	-8 + 16	5.46	-	-
-S2	22.84	1015	-16 + 20	7.60	9.07	10.28
-S3	46.24	630	-20 + 40	8.58	9.61	10.75
-S4	12.17	335	-40 + 60	9.40	11.55	11.99
-S5	6.18	213	-60 + 80	10.87	11.28	12.14
-S6	3.09	163	-80 + 100	11.06	11.90	-
-S7	0.84	137	-100 + 120	-	-	11.70
-S8	1.17	127	-120 + 140	11.04	-	-
-S9	0.83	115	-140 + 160	-	11.02	-
-S10	0.39	100	-160 + 180	-	-	11.61[c]
-S11	0.40	77.5	-180 + 200	-	-	
-S12	4.54	55	-200	11.25	11.25	-

a Milled on No. 2 Mill with Nylon guides at 0.176 inch spacing to net input powers between 1.15 and 1.18 KW.

b Ground with Cumberland grinder using a 1/16-inch screen, classified with Rotatap.

c Blend of S-10 and S-11.

A comparison with last month's report shows that material ground with the Cumberland grinder contains less fines than material ground in the swing-sledge grinder. The finer screens of the Cumberland also materially reduce the number of large particles which stabilize much more slowly.

Removal of the fines from plastic for production molding should not result in an excessive loss when the Cumberland grinder is used.

The immediate problem is to determine the net input power to which our formulations should be milled to give a product which is optimum for use by production molding. The correlation of net input power with cup-flow test data can be completed at a later date.

Plastic Evaluation -Test specimens were molded from a number of DAP formulations. None of the moldings could be considered excellent, but a few were better than most test specimens made during the last few months. Heat-distortion temperature determinations were made on all but the two for which usable test bars could not be obtained. Izod impact strength measurements were made on all, even though most of the test specimens probably contained internal cracks. The large number of bad breaks obtained in the tensile strength tests is an indication of the questionable nature of the moldings. The test data are summarized in Table XI.

TABLE XI

Physical Properties of DAP Formulations

Sample Number	Remarks on Tensile Measurement Breaks	Tensile Strength lb/inch^2	Izod Impact Strength ft-lb/in. of notch	Heat Distortion Temperature °C
B20-8	Tensile and Impact data for B20-8 Through B28 from previous report, all test specimens were poor moldings.	6700	0.224	145
B25		4670	0.253	163.5
B26		4000	0.265	152
B27		4800	0.245	144
B28		4800	0.245	139
B29-9		-	0.259	151
B30-1	All 3 bad	3720	0.257	154
B30-5	1 good, 2 bad, bad higher	4407	0.264	138
B30-7	All 3 bad	5082	0.241	140
B31-1	1 bad, 2 questionable	3811	0.244[a]	-
32-1	All 3 good	5527	0.256	143
32-6	All 3 good	6027	0.239	144
B4-1/5 S1	2 good, 1 questionable	4402	0.264[a]	-
35-6	-	-	0.261	161
Mesa B-1143	3 good breaks	5630	0.256	156
		-	—	—
Mesa B-1310	4 good breaks	6370	0.270	159

a Moldings very poor only one sample run, Average of four breaks for all other impact strength determinations.

The poor quality of the test specimen moldings was due in part to the fact that most of the samples were quite stiff. However, all samples gave good moldings in the cup-test mold. The major difficulty, therefore, appears to be the long times required to close the Wabash press and to develop molding pressure. The plastic is partially cured before full molding pressure can be applied. Increase in the length of stroke for the motorized drive has shortened the time required to develop pressure after closing, while the application of 100 psi air pressure to the top of the oil reservoir permits the closing of the press in about four seconds.

Ten preforms were made from B35-6 for test molding by Production Molding. Five of the 50 heads molded from this material were used for chloroform extraction and gave an average cure-time of 1.3 minutes. The remaining 45 were X-rayed, and 23 were rejected for cracks, 3 for voids, and 2 for inclusions. It is believed that the high percentage of cracks was caused by the fact that B35-6 has a high value of ultimate flow. There was a marked reduction in the size and number of cracks per head and in the percentage of rejects for cracks when compared to the heads molded previously from material known to be stiffer than B35-6.

Additional attempts have been made to determine if heat is evolved during the aging of DAP formulations. The results to date are inconclusive. There are definite indications of heat evoluation, but the quantity of heat evolved per unit time has been so small that it cannot be stated positively that these indications are

truly the result of evolution of heat by the sample There is some evidence that the very constant temperature maintained in the calorimeter may retard the aging process; if so, the rate of heat evolution in the calorimeter would be much less than for the same sample under normal room conditions.

Production Material Control The experiment with the spring-loaded knock-out pins was inconclusive The possible advantages appear to be outweighed by the fact that when the press is opened and the parts are knocked out, the mold cavity block is very difficult to remove from the press The spring tension on the mold cavity block causes it to cock and bind in the mold chase.

The redesigned transfer plunger has been used for a sufficient number of moldings to indicate that it has definite advantages over the transfer plunger presently used

Work has continued with the Production and Engineering Divisions on the design for the 4E8 mold A preliminary work order request has been initiated by the Production Division for the design of a new mold for the 1E26. This will incorporate our ideas for a split cavity which we believe will minimize several present difficulties in production molding

GENERAL WEAPONS DEVELOPMENT

Process development work is being conducted on the use of thermal diffusion for the production of certain gaseous isotopes which are not at this time identified with particular weapons, but which are needed for research and development purposes in the weapons program.

Isotope Separation Project

Gaseous Thermal Diffusion Column Operation - A study was made of the effect of relative concentration of gases in feed mixtures on the thermal diffusion column separation factors. Mixtures of hydrogen and helium and mixtures of hydrogen and deuterium were used in the 1.61-inch column with a molybdenum center-wire 0.064-inch in diameter. The data obtained showed that the separation factor of a continuous-flow system could be affected seriously by the feed composition. For hydrogen-helium mixtures, the separation factor decreases with increased hydrogen concentration in the feed. For hydrogen-deuterium mixtures the separation factor increases with increased hydrogen concentration. Evaluation of these data is still in progress.

Work has started on the determination of operating parameters for the separation of oxygen isotopes by thermal diffusion. Since molybdenum oxidizes readily at high temperatures, other materials were examined for suitability as center-wires in an oxygen environment. Nichrome V was selected for center-wire material. Two Nichrome V center-wires have been in use approximately 300 hours at 900°C in pure oxygen without visible signs of deterioration. Wires that were sufficiently straight for use in the thermal diffusion columns were obtained by stretching a length of the wire and annealing it for a few hours in air.

Closed-end operation of the 1.61 and 1.50-inch diameter columns in cascade at the mean optimum pressure of approximately 110 millimeters of mercury with center-wire temperatures of 900°C gave a separation factor between oxygen-16 and oxygen-18 of about four. After approximately one day of continuous draw-off at the top of the cascade with the feed port for oxygen of normal abundance two feet below the top of the twenty-four foot cascade, a 20 milliliter sample containing approximately 1 per cent oxygen-18 was obtained from the bottom of the cascade.

Work is now in progress to connect the 1.25 and 1.00-inch columns in cascade with the 1.61 and 1.50-inch columns. The four-column cascade will be used for further studies on separation of oxygen isotopes.

Six hydrogen and helium samples and ninety oxygen samples were analyzed on the mass spectrometer this month. This work was done in conjunction with the above experimentation.

Gas Purification - Installation of the gas purification line in R-106 was completed, with the exception of the palladium diffuser, and pressure-tested with helium. The palladium diffuser showed leakage and has been temporarily by-passed.

Several leaks in the silver-soldered tubing connections were found and repaired. The Teflon-gasketed flanged joints for one of the tube furnaces did not seal, so the flanges were modified to increase the sealing area. Leaks developed through the threads of the standard pipe fittings for one of the oil filters, and these have been repaired by silver soldering the threaded joints. The metal surfaces next to the Teflon gaskets on the same filter were found to be rough, which caused a poor seal. These surfaces have been smoothed.

Final design and drawings were completed for the system to be installed in R-105 for the purpose of pumping by-product gas from the trailer tanks into propane tanks for feeding the gas purification line. All equipment is on hand for this pumping system except a Copelmatic Compressor, which is on order.

Liquid Thermal Diffusion - Experimental runs using the 50-50 volume per cent benzene-carbon tetrachloride system as feed have been continued to obtain operating experience. Accumulation of gas at the top of the annulus appears to have been minimized by first degassing the feed by boiling gently in vacuo for several minutes, then filling the evacuated annulus through the bottom port. During the run, the feed line is left open to the middle port.

Table I shows the per cent separation obtained at various time intervals, based on various pairs of fractions. The nominal six-foot high annulus is divided into nine fractions, and these are numbered consecutively from bottom to top. Benzene tends to concentrate in the top fractions and carbon tetrachloride in the bottom fractions. The per cent separation is defined as the difference in refractive indices of two fractions expressed as per cent of the difference from the refractive indices of the pure components.

TABLE I

Effect of Time on Per Cent Separation for Different Pairs of Fractions

Feed: 50-50 volume per cent C_6H_6 - CCl_4
Hot Wall: 50°C
Cold Wall: 17°C

Time Hr	Per Cent Separation Fractions 4 and 6	Fractions 3 and 7	Fractions 2 and 8	Fractions 1 and 9
3	13.1	25.9	46.9	74.0
4	17.3	35.6	58.5	81.5
7.5	39.6	64.1	82.1	90.8
7.5	42.2	69.1	84.8	90.1
24	87.1	96.7	97.8	98.6
24	94.0	99.0	99.6	99.5
48	96.5	99.1	99.6	99.9
48	96.5	98.9	99.4	99.6
48	92.6	96.8	99.3	99.3

The per cent separation reported in the second column of the table is based on the difference between the refractive indices of fractions 4 and 6 which are situated immediately below and above, respectively, the feed fraction (fraction 5). The third column gives the per cent separation based on the difference between fractions 3 and 7, each of which is two fractions removed from the feed fraction; the fourth column gives the separation based on fractions 2 and 8, each of which is three fractions removed from the feed fraction; and the last column gives the separation obtained with the end fractions. In all cases, the hot wall was at approximately 50°C and the cold wall was at approximately 17°C.

In the 48-hour runs about 99.5 per cent separation between the two end fractions as well as between the two fractions once removed from the ends was obtained. Upon receding further from the ends, the separation dropped off somewhat, becoming perhaps 95 per cent between the fractions adjacent to the feed fraction. The 24-hour runs gave essentially the same results as the 48-hour runs except for a possible minor reduction in separation for the fractions adjacent to the feed fraction. Thus 24 hours seems to be nearly adequate for establishment of equilibrium concentractions in the column. Further reduction in time resulted in marked decreases in separation which became increasingly evident in fractions nearer the feed fraction.

The installation of the second liquid thermal diffusion column has been completed except for thermocouple connections. Special pycnometers have been received, and a thermostat for use in density determinations will be installed shortly.

REACTOR DEVELOPMENT PROGRAM

CIVILIAN POWER REACTORS

HOMOGENEOUS REACTOR

The Civilian Power Reactor Program is concerned with the development of reactors suitable for the production of power or heat for civilian use. Homogeneous reactors are one of the types being investigated. In a homogeneous reactor, the fuel is by definition evenly dispersed in the moderator, so that solutions of uranium-233, uranium-235 or plutonium-239 are acceptable fuels if their chemical and physical properties permit low capital and operating costs.

Plutonium Solubility Studies

A project has been initiated to investigate aqueous systems containing plutonium as a fuel for a homogeneous reactor. Initially the solubility of various plutonium salts in aqueous solution up to 300°C will be measured. The maximum concentration of plutonium will be ten grams per liter. A second solubility study will be carried out on an aqueous system containing up to 300 grams of uranium per liter and five grams of plutonium per liter.

Two series of determinations were made on the solubility of the system plutonium (IV) sulfate-sulfuric acid-water. The results are listed in Table I.

TABLE I

Plutonium Sulfate Solubility in Sulfuric Acid-Water Systems

Plutonium (IV) Sulfate in 2*N* H_2SO_4		Plutonium (IV) Sulfate in 2.57*N* H_2SO_4		Plutonium (III) Sulfate in 0.067*N* H_2SO_4	
Pu Conc. g/liter	Temp of Phase Change °C	Pu Conc. g/liter	Temp of Phase Change °C	Pu Conc. g/liter	Temp of Phase Change °C
12.45	254.3	13.18	257.7	13.28	166.2
8.98	253.4	8.43	254.3	8.57	174.8
4.98	248.0	5.34	256.6	5.69	197.2
1.52	> 340.0	3.35	264.6	3.36	204.8
1.09	> 341.5	1.72	> 337.0	1.81	233.2
0.68	> 339.5	1.35	> 338.2	1.14	249.0
		0.91	> 338.0	0 76	262.0

These runs complete studies on this system, and a summary of the results is given in Figure 1. One series of runs was made on the system plutonium (III) sulfate-sulfuric acid-water. Results are given in the table.

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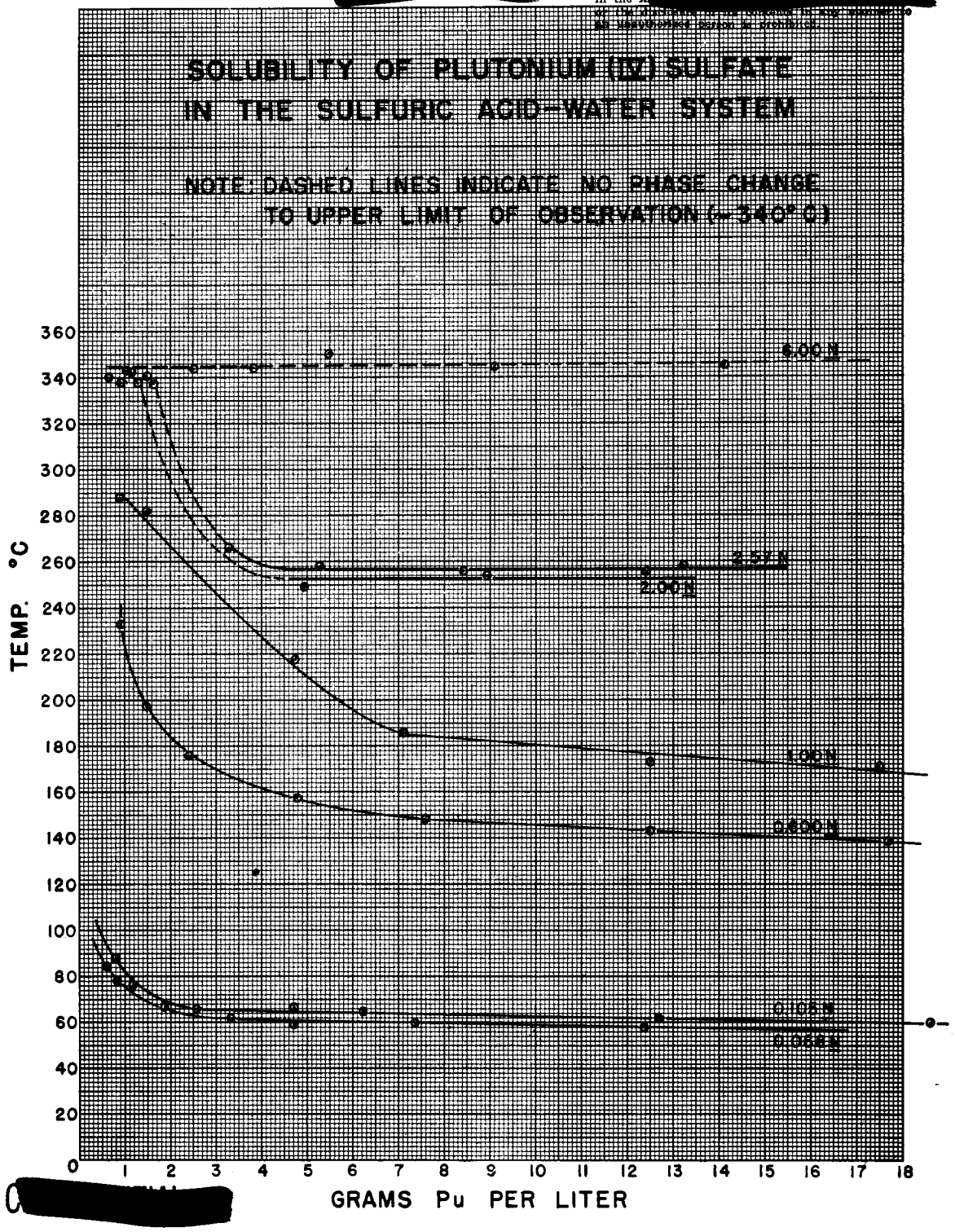

FIGURE 1

[redacted]

The phase change that occurred was the appearance of a well-formed, long, light blue crystal quite in in contrast to the muddy, granular hydrate formed in the plutonium (IV) sulfate solutions.

Plutonium (III) sulfate was prepared in a closed system by reduction of plutonium (IV) sulfate in dilute sulfuric acid with sulfur dioxide. The solution was evaporated and the crystals were washed with methyl alcohol twice, dissolved in the desired concentration of sulfuric acid, and stored under a very slight sulfur dioxide atmosphere to prevent reoxidation. Titration indicated that 100 per cent of the plutonium was in the trivalent state.

AIRCRAFT PROPULSION REACTORS

LIQUID CYCLE REACTORS

The Aircraft Propulsion Reactors activity is concerned with the development of reactors suitable for the propulsion of aircraft or missiles, or suitable for the production of auxiliary power for such vehicles. A liquid-cycle reactor is one of the types under development. In such a reactor, a liquid containing the fuel circulates through the reactor and a heat exchanger. The fluid for operating a turbine or jet is heated at the exchanger. Suitability of liquids for such reactors is based on extensive knowledge of their physical and chemical properties.

Fused Salts Research Project

The Liquid Cycle Reactor is considering the use of a fused-salt fuel system. Mound Laboratory has been assigned the problem of determining the phase relationships and physical properties of the components of some of the proposed fuel systems.

LiF-RbF-BeF_2 Ternary System - Data obtained to date from the differential thermal analysis studies of mixtures of LiF, RbF, and BeF_2 are summarized on Figure 1. These data include the thirty determinations made during the past month.

A partial phase and temperature contour diagram showing the interpretation of data obtained up to the present time appears as Figure 2. In addition to the invariant points on this diagram, five unassigned transitions at 315°, 320°, 425°, 466°, and 518°C have been observed during differential thermal analyses of various compositions in the uncompleted area of this diagram. These transitions will be designated either eutectics or peritectics as soon as other compositions are examined and the data obtained are evaluated. The appearance of these transitions during differential thermal analyses indicates existence of one or two ternary compounds in this system

Differential thermal analysis studies have established a quasi-binary between LiF and Rb_2BeF_4 with a eutectic at 640°C having a composition of 37 mole per cent LiF, 42 mole per cent RbF, and 21 mole per cent BeF_2. There is no quasi-binary between LiF and Rb_3BeF_5.

Upon cooling from above the liquidus temperature the course of crystallization of ternary compositions within the LiF-Rb_2BeF_4-Rb_3BeF_5 composition triangle stops at a 527°C peritectic having an approximate composition of 41 mole per cent LiF, 52 mole per cent RbF, and 7 mole per cent BeF_2, and located outside of this composition triangle as indicated in Figure 2

A eutectic composition whose melting point is 462°C and whose composition is approximately 43 mole per cent LiF, 55 mole per cent RbF, and 2 mole per cent BeF_2 has been found immediately above the eutectic point on the LiF-RbF binary system.

The lithium- and rubidium-rich portions of this ternary system are very adaptable to static examination during differential thermal analyses. Curves obtained during both the heating and cooling cycles yield reliable data without agitation of the sample. Because of the relatively high temperatures, low viscosities, and rapid equilibrium rates prevailing in the lithium- and rubidium-rich areas of this ternary system, the thermal effects due to peritectic transitions are easily observed. However, rupture of the sample capsules containing ternary compositions on or below the LiF-Rb_2BeF_4 quasi-binary line during the first or second heating cycle at or immediately above the liquidus temperature has given rise to a difficult problem.

LIQUIDUS AND EUTECTIC TEMPERATURES (°C)
OBTAINED BY DIFFERENTIAL THERMAL ANALYSIS
FOR SYSTEM LiF–RbF–BeF_2

TOP NUMBER – LIQUIDUS
BOTTOM NUMBER – EUTECTIC

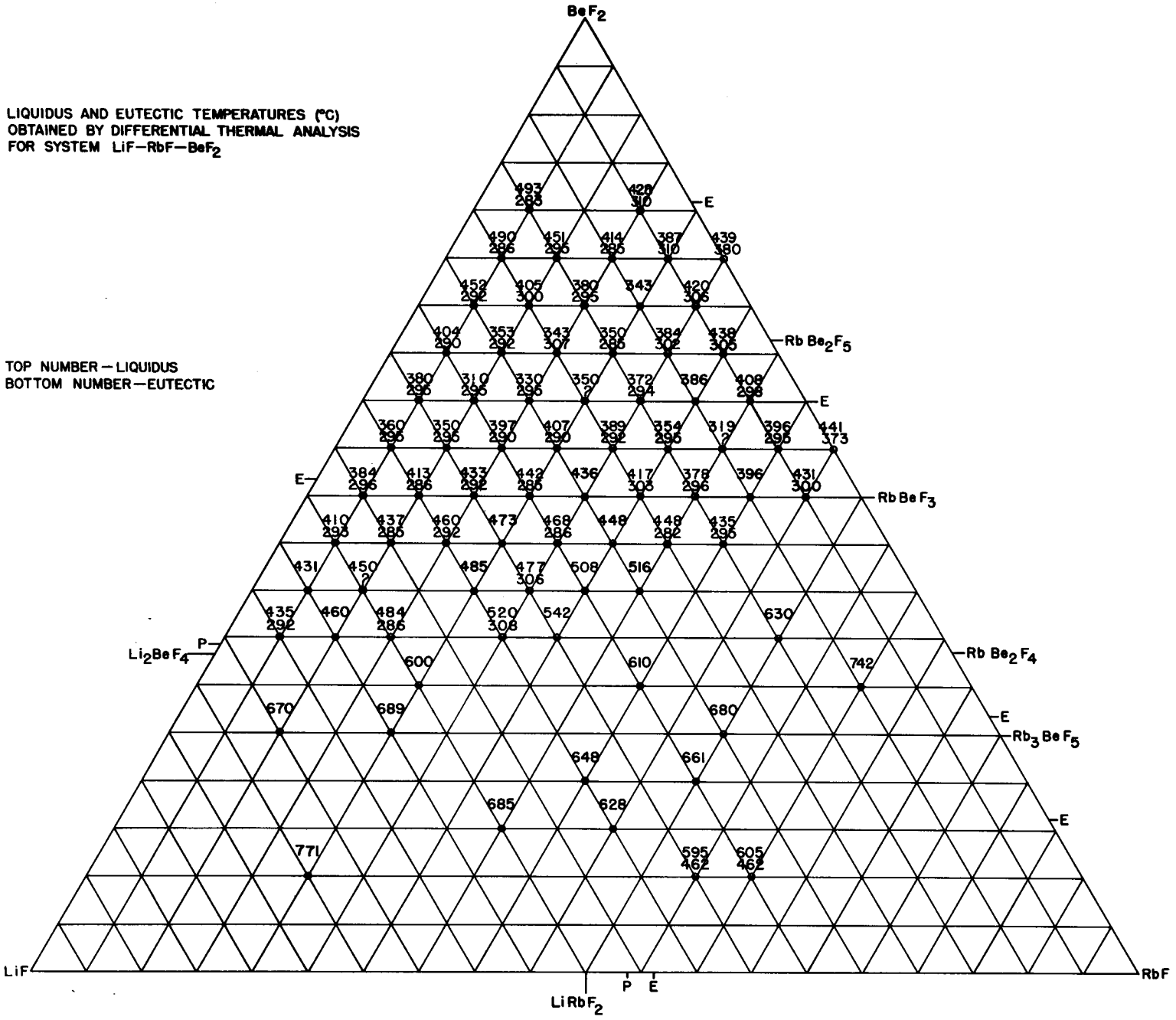

FIGURE 1

Only one or, at the most, two runs can be made on any one sample of this type. The capsule breakdowns have been attributed to a combination of the high temperatures and the presence of rubidium fluoride. This problem was not experienced during the examination of the LiF-BeF_2 binary system.

A small primary phase area in this ternary system for $RbBeF_3$ has been established. The $RbBeF_3$ was identified by petrographic analysis as the primary phase from compositions R-22 and R-23, listed in Table I. This identification was verified by X-ray diffraction techniques.

TABLE I

Thermal-Gradient Quenching Data of the LiF-RbF-BeF_2 System

Sample No.	Composition Mole Per Cent LiF	RbF	BeF_2	Phase Change Temp. °C	Phases Found Just Above Phase Change	Phases Found Just Below Phase Change	Interpretation
R-22	5	42	53	>378	Liquid	Liquid, $RbBeF_3$	Liquidus and primary phase
				352	Liquid, $RbBeF_3$	Liquid, $RbBeF_3$ $RbBe_2F_5$	Boundary intersection
R-23	10	35	55	314	Liquid	Liquid, $RbBeF_3$	Liquidus and primary phase
				308	Liquid, $RbBeF_3$	Liquid, $RbBeF_3$ $RbBe_2F_5$	Boundary intersection
R-24	15	46	39	~566	Liquid	Liquid, Rb_2BeF_4	Liquidus and primary phase
				~480	Liquid, Rb_2BeF_4	Liquid, Rb_2BeF_4, Comp. B	Boundary intersection
R-25	33.3	33.3	33.3	>568	Liquid	Liquid, LiF	Liquidus and primary phase
				542	Liquid, LiF	LiF, Comp. B	Solidus at the boundary intersection
R-28	45	18	37	~473	Liquid	Liquid, LiF Comp. A	Liquidus and composition point on boundary
R-29	35	20	45	~482	Liquid	Liquid, Comp. A	Liquidus and primary phase
R-30	27	30	43	458	Liquid	Liquid, Comp. B	Liquidus and primary phase

Since $RbBeF_3$ exists as a primary phase in the ternary system, it is believed that the two different X-ray patterns which have been repeatedly obtained from various compositions in the middle areas of the ternary system are the patterns of two different ternary compounds. The ternary phase areas for these compounds appear to be adjacent. The optical properties of Compound A and Compound B are practically

[redacted]

identical to those of $RbBeF_3$ so that identification by petrographic techniques is impossible. X-ray diffraction examinations have established the existence of Compounds A and B. Current X-ray diffraction information suggests that Compound B has the empirical formula, $LiRbBeF_4$. The primary phase areas of these two compounds cannot be established until additional data have been obtained to establish the course of the boundary lines and the locations of the invariant points.

The ternary composition having a ratio of components of 1:1:1 (R-25) was examined by petrographic techniques to determine whether or not it represented the composition point of Compound B. LiF appeared to be the primary phase. The solidus temperature was approximately 540°C. At this temperature only LiF and Compound B were found. Compound B apparently is on the LiF-$RbBeF_3$ join and melts incongruently to yield LiF.

High-Temperature Calorimetry - Twenty-six simultaneous determinations of the heat capacity and thermal conductivity of Composition No. 30 were made at temperatures between 40° and 612°C. The heat of fusion and the temperature interval in which melting occurs were also determined. The sample, consisting of 44.0 grams of Composition No. 30, was prepared at this laboratory and transferred into the thermal conductivity cell by pouring the molten mixture. The conductivity cell used for these measurements was Cell No. 2, described in the previous Monthly Technical Activities Report (CF-57-4-32). The cell was filled to within one-quarter inch of its top and was sealed by welding.

This conductivity cell had been previously checked with water and good agreement for the thermal conductivity of water with published values was obtained. The performance of the apparatus at temperatures up to about 620°C was good. The precision of the data obtained remained essentially constant over the termperature range investigated. The accuracy of the thermal conductivity measurements cannot be completely evaluated at this time. The defining equation given in the previous report (CF 57-4-32) was not complete for the situation evaluated. If the thermal resistance between the outer surface of the conductivity cell and the bridge winding of the calorimeter is considered, the equation becomes:

$$K_S = \frac{\frac{C_S(r_1^2-r_2^2)}{4} + \frac{(C_n-C_S)\, r_2^2 \ln(\frac{r_1}{r_2})}{2}}{\frac{A}{T_1-T_2} + \frac{C_n(r_1^2-r_2^2)}{4K_n} + \pi (C_n-C_S)(r_1^2-r_2^2) R}$$

in which R (°C-sec-cal^{-1}-cm) is the thermal resistance per unit length from the outer surface of the conductivity cell to the bridge of the calorimeter. The opposite side of the calorimeter (dummy side) is assumed to have the same value for R. In the case of water, the value of C_n-C_S is extremely small, so this entire term becomes negligible. However, for Composition No. 30 the value of C_n-C_S is appreciable, particularly near the Curie point of nickel where $C_n > C_S$. In the temperature interval where melting is taking place, $C_S \gg C_n$. Since this thermal resistance R, cannot be eliminated, it must be measured. The value of R will not change if different cells are used, providing the outside dimensions of the cells remain constant. However its value is temperature-dependent. Since no subsolidus transitions occur in Composition No. 30, they cannot be used to evaluate this term. The melting point cannot be readily used for this purpose since a change in volume of the salt system usually occurs during the melting process.

In order to evaluate this thermal resistance, R, a second thermocouple is being added to the thermal conductivity cell. One thermocouple will be in the center of the cell as before, and the second will be in the outer wall of the cell. The wall thickness has been increased from 0.03 inch to 0.13 inch.

[redacted]

The complete results obtained from the thermal measurements made to date are listed in Table II. The values for the thermal conductivity of Composition No. 30 are given as uncorrected for the effect of R and as tentatively corrected assuming a linear change for R from 200°C-sec-cal^{-1}-cm at 40°C to 110 °C-sec-cal^{-1}-cm at 520°C.

Calculated heat capacities are plotted in Figure 3. Acceptable agreement is indicated between the values obtained by Powers and Blalock at Oak Ridge National Laboratory and the values obtained at this Laboratory.

TABLE II

Thermal Measurements on Composition No. 30

Temperature Interval of Measurement °C		Heat Capacity cal-g^{-1}-°C^{-1}	Thermal Conductivity (K^1) (Uncorrected) cal-°C^{-1}-sec^{-1}-cm^{-1}	Thermal Conductivity (K) (Corrected) cal-°C^{-1}-sec^{-1}-cm^{-1}
T_2	T_1			
30.50	48.88	0.174	0.0046	0.0024
48.88	100.04	0.178	0.0047	0.0024
27.64	100.40	0.179	0.0053	0.0025
100.40	201.88	0.185	0.0059	0.0027
201.88	300.70	0.188	0.0136	0.0033
450.00	503.55	0.218	0.0084	0.0048
503.55	523.87	3.055	0.00087	0.0039
523.87	543.87	0.382	0.0024	0.0029
28.23	125.03	0.177	0.0079	0.0030
125.03	226.05	0.183	0.0083	0.0029
226.05	326.28	0.196	0.0123	0.0036
326.28	426.37	0.207	0.0130	0.0043
426.30	465.31	0.206	0.0082	0.0041
465.31	503.46	0.249	0.0088	0.0065
505.39	514.88	3.330	0.00088	0.0046
514.88	518.56	4.026	0.00091	0.0062
518.56	522.11	2.666	0.00080	0.0026
522.11	533.26	0.778	0.0011	0.0020
533.24	552.38	0.288	0.0041	0.0035
552.38	572.35	0.278	0.0043	0.0034
572.35	592.34	0.276	0.0044	0.0035
592.34	632.29	0.266	0.0048	0.0035
375.23	426.06	0.209	0.0064	0.0036
426.06	465.12	0.211	0.0067	0.0038
465.12	485.17	0.216	0.0065	0.0039
485.17	505.18	0.225	0.0064	0.0042

Density and Viscosity - The density and viscosity data recently obtained from various quaternary mixtures of LiF, NaF, BeF_2 and UF_4 are reported in Table III. All reported data were obtained between the liquidus point and 900°C. Interpolated values are given at 800 and 700°C and at 600°C when possible.

[redacted]

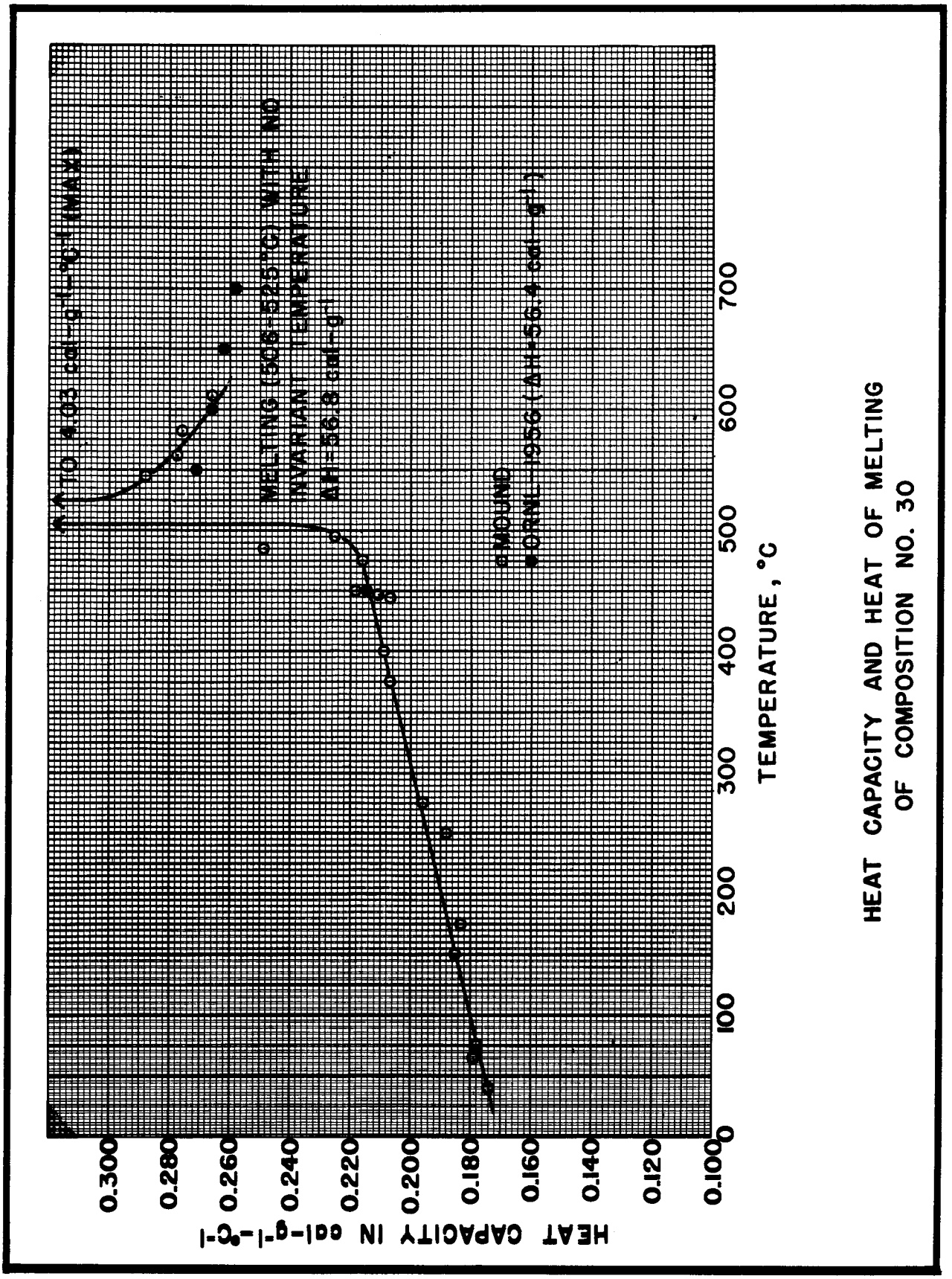
HEAT CAPACITY AND HEAT OF MELTING
OF COMPOSITION NO. 30
TEMPERATURE, °C
HEAT CAPACITY IN cal-g⁻¹-°C⁻¹
0.300
0.280
0.260
0.240
0.220
0.200
0.180
0.160
0.140
0.120
0.100
0
100
200
300
400
500
600
700
MELTING (506-525 °C) WITH NO
INVARIANT TEMPERATURE
ΔH = 56.3 cal-g⁻¹
←TO ± 0.3 cal-g⁻¹-°C⁻¹ (MAX)
● ORNL - 956 (ΔH=56.4 cal-g⁻¹)
○ MOUND

FIGURE 3

TABLE III

Density and Viscosity of Mixtures of UF_4, NaF, LiF and BeF_2

Composition Mole Per Cent UF_4	NaF	LiF	BeF_2	Melt. Point °C	600°C η Poise	600°C ρ g/cm³	600°C η/ρ Stokes	700°C η Poise	700°C ρ g/cm³	700°C η/ρ Stokes	800°C η Poise	800°C ρ g/cm³	800°C η/ρ Stokes
6	13	34	47	345	0.2400	2.492	0.0963	0.1120	2.441	0.0459	0.0585	2.430	0.0241
6	25	22	47	417	0.2040	2.556	0.0798	0.0990	2.503	0.0396	0.0540	2.350	0.0220
6	35	12	47	394	0.1870	2.553	0.0732	0.0853	2.493	0.0342	0.0450	2.480	0.0181
6	46	12	36	402	0.1030	2.626	0.0392	0.0560	2.562	0.0219	0.0334	2.497	0.0134
6	70	12	12	651	-	-	-	0.0613	2.657	0.0231	0.0412	2.607	0.0158
6	70	24	0	733	-	-	-	-	-	-	0.0350	2.683	0.0130
6	46	48	0	580	0.0568	2.796	0.0203	0.0368	2.726	0.0135	0.0256	2.655	0.0096

The measurement of the densities and viscosities of this quaternary system has been terminated at a maximum of 6 mole per cent uranium tetrafluoride.

The data listed in Table IV are the results of three of the twelve density measurements which were re-examinations of systems previously measured. Values obtained from these three reruns were essentially different from the density information previously reported. During the density measurements, the formation of an insoluble solid, possibly due to oxide formation, above 850°C in the three systems reported caused some uncertainty.

TABLE IV

Density and Viscosity of Mixtures of UF_4, NaF, LiF and BeF_2
Reruns

Composition Mole Per Cent UF_4	NaF	LiF	BeF_2	Melt. Point °C	600°C η Poise	600°C ρ g/cm³	600°C η/ρ Stokes	700°C η Poise	700°C ρ g/cm³	700°C η/ρ Stokes	800°C η Poise	800°C ρ g/cm³	800°C η/ρ Stokes
4	46	50	0	604*	-	-	-	0.0577	2.432	0.0237	0.0431	2.382	0.0181
4	35	26	35	315*	0.0997	2.446	0.0408	0.0570	2.392	0.0238	0.0357	2.339	0.0153
2	35	32	31	340*	0.1065	2.259	0.0471	0.0652	2.203	0.0296	0.0432	2.149	0.0201

* Salt build-up.

[redacted]

DEPARTMENT OF DEFENSE

U. S. ARMY SIGNAL ENGINEERING LABORATORIES

Ft. Monmouth

COMPONENTS BRANCH

(Contract No. R-50-799965-SC-01-91)

Nuclear Battery - Thermoelectric Type

The Army Signal Corps Engineering Laboratory is concerned with the development of sources of electricity with particular application to military use. They are interested in a battery suitable as a power source of moderate output, light weight, small bulk and ability to operate under extreme conditions.

The development of a thermal energy conversion system for the direct conversion of the energy of radioactive decay as heat into electrical energy is under investigation by Mound Laboratory. Radioactive materials will be evaluated as heat sources with consideration given to abailability, cost, half life, shielding, health hazard and efficiency. Thermocouples and factors for optimum shape and configuration for maximum efficiency output will be considered and prototype thermal battery will be constructed.

An error was found in the factor used in calculating the watt-hour per year values for isotopic heat sources. Corrected values are given in Table I.

Consideration of a number of isotopes as heat source possibilities for the battery was discontinued. There are not sufficient data upon which to base further calculations for argon-42, europium-149, osmium-194, and tungsten-188. Promethium-146, gadolinium-148, lutetium-174, lead-210, polonium-208, polonium-209, and curium-242 do not have any economical production schemes. Nickel-63 and samarium-151 do not produce the minimum of 10^4 watts per mole. Krypton-85 was eliminated because no method could be visualized for containing sufficient compressed gas to form a heat source. The following isotopes and combinations remain: sulfur-35, calcium-45, nickel-63, strontium-89, strontium-89 + strontium-90 +yttrium-90, strontium-90 + yttrium-90, yttrium-91, cadmium-113m, tin-121m + tin-123, tin-123, cerium-144 + praseodymium-144, promethium-147 samarium-151, europium-152 + europium-154, europium-155, thulium-170 +thulium-171 (from thulium-169), thulium-171 (from erbium-170), tungsten-185, thallium-204, and polonium-210.

The power per cubic centimeter of each isotope has been calculated, and source-size and temperature calculations were begun.

TABLE I

Power per Mole for Various Isotopic Heat Sources

Isotope	Energy per Mole of Pure Isotope per Year	Isotope	Energy Per Mole of Pure Isotope per Year
sulfur-35	1.69×10^6	europium-152	1.00×10^6
argon-42		europium-154	6.25×10^5
calcium-45	2.14×10^6	europium-155	6.29×10^5
nickel-63	5.82×10^3	gadolinium-148	4.15×10^5
krypton-85	4.66×10^6	thulium-170	8.76×10^6
strontium-89	1.56×10^7	thulium-171	3.37×10^5
strontium-90	7.46×10^5	lutetium-174	1.01×10^6
yttrium-91	1.64×10^7	tungsten-185	4.46×10^6
cadmium-113m	7.89×10^5	tungsten-188	–
tin-121m	5.80×10^5	osmium-194	–
tin-123	1.30×10^7	thallium-204	1.64×10^6
cerium-144	1.99×10^7	lead-210	5.43×10^6
promethium-146	3.75×10^6	polonium-208	2.88×10^7
promethium-147	5.73×10^5	polonium-209	8.11×10^5
samarium-151	5.55×10^3	polonium-210	1.19×10^8
		curium-244	1.29×10^8