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MOUND LABORATORY

Operated By

MONSANTO CHEMICAL COMPANY

MIAMISBURG, OHIO

MONTHLY TECHNICAL ACTIVITIES REPORT THROUGH JUNE 15, 1957

By

J. F. Eichelberger and D. L. Scott

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Excluded from automatic downgrading and declassification

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Date: June 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.

Eleven polonium-beryllium neutron sources, two alpha sources, and two plutonium-beryllium neutron sources were shipped in May. Six polonium-beryllium neutron sources, one alpha source, and one plutonium-beryllium source were for other AEC sites.

Twenty-six tanks of influent water were processed during this period. Two of the tanks required no pretreatment. Eight tanks required extensive treatment with barium chloride and sodium sulfide.

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

TABLE I

Discharge Volume	711,600	gallons
Total Alpha Activity	11.0	millicuries
Total Beta Activity	30.1	millicuries
Activity Density		
Alpha	4.5	cts/min/ml
Beta	12.4	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.

Ten drums of sulfated feed (Raffinate AM-8) from Mallinckrodt Chemical Works were moved into the R-Building high-risk corridor for separation of uranium from thorium before processing in the mixer-settler units. This preliminary separation was done to take advantage of the low distribution coefficient of thorium in the sulfated solution and to remove some of the excess uranium which this feed contained. Shipping data from Mallinckrodt clearly indicated that 1334.5 pounds of feed were shipped in ten 15-gallon drums, and contained a total of 16 kilograms of uranium, one kilogram of thorium, and 129 grams of ionium.

The volume was estimated to be 134.11 gallons from measurements in the drums as received. From the density, found to be 9.693 pounds per gallon, and the total weight shipped, the volume was calculated to be 137.68 gallons. However, the volume found after the feed solution had been emptied into process tanks was 133.91 gallons. The assay of this feed solution was 28.1 grams per liter of uranium, 2.6 grams per liter of thorium, and 0.218 grams per liter of ionium. The solution was 0.86-normal in nitric acid. The uranium and thorium were determined by X-ray fluorescence using yttrium as the internal standard. Ionium analysis was by alpha counting. Calculations indicated only one alpha from Th-232, U-238, and U-234 would be present for every 1000 alphas from Th-230. An alpha spectrum of the feed solution corroborated these calculations. On the basis of these analyses, a total of 14.24 kilograms of uranium, 1.32 kilograms of total thorium, and 110.5 grams of ionium was received.

A dummy feed solution was prepared with 32.2 grams of uranium per liter, and 3.3 grams of natural thorium per liter, and was one normal in nitric acid and one molar in sodium sulfate. Extraction data were gathered using varying ratios of the 40-60 TBP-Amsco extractant to the aqueous feed over a range from 1 to 2.5-normal nitric acid and also from 0.5 to 2.0-normal aluminum nitrate in addition to the nitric acid. These experiments showed each ratio of feed to organic extractant approached a peak separation of uranium and thorium with the increase of the normality of the solution and then dropped off. They also showed, as expected, that the separation of uranium and thorium increased as the volume of the organic extractant was decreased.

Maximum separation was obtained at 1.82-normal nitric acid using three parts of feed to one part of organic. The organic phase contained 50.4 per cent of the total uranium and 5.0 per cent of the total thorium, giving a ratio of uranium to thorium of 10.1 to 1. However, with 2.14-normal nitric acid, and using two parts of feed to one part of organic, 67.0 per cent of the total uranium and 7.3 per cent of the total thorium was extracted, giving a uranium-thorium ratio of 9.2 to 1. The latter conditions were chosen since less of the uranium would be present when the feed is processed in the mixer-settler units.

The 134 gallons of feed were processed as outlined above in six batches with fifteen minutes agitation of the feed with the organic. After thirty minutes of settling, the feed was separated from the organic. The organic phase was neutralized with solid sodium carbonate and agitated with approximately four parts organic to three parts by volume of ten per cent sodium carbonate solution. All of the uranium and thorium was removed from the organic, and the organic was re-used for each batch extraction. Analysis showed the treated feed still contained 23.8 per cent of the total uranium and 96.5 per cent of the total thorium. Experiments are being conducted to increase the distribution coefficient of thorium to a value high enough to process in the mixer-settler units by using aluminum nitrate.

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 - All 23 drums of the original Sperry Cake have been digested and processed. In addition, the 14 drums that were partially processed by the hydrochloric acid head-end treatment have been digested and are presently being processed. This material, which is the remaining half of the original 80 drums of Sperry Cake, seemed to contain more iron, and the per-cent of protactinium extracted was lower than anticipated. The raffinates from this processing have been saved, and a method is now being developed to extract the remaining protactinium, which amounts to approximately 40 per cent of that originally present. Preliminary tests indicate that the addition of hydrochloric acid will contribute to a good protactinium extraction from these raffinates. The iron was nearly all removed in the first processing, so it should not be present in sufficient quantity to interfere with the re-processing of the raffinates.

Table I is the best available estimate of the progress of the protactinium separation. It is estimated that the data in the table could be in error by as much as 20 per cent.

TABLE I

Protactinium Processing Data

Run No.	Sperry-Cake Drum Number	Protactinium Available[a] mg	Protactinium Unextracted %	Total Protactinium Recovered mg
HH-1	45,46,66	34	45	19
HH-2	49,57,63,68	57	88	7
HH-3	41,47,54,67	125	52	60
HH-4	55,56,65,80	88	52	42
HH-5	44,48,64,78	83	41	49
HH-6	50,58,69,79	68	41	40
HH-7	All partially processed drums	367	17	305
Total		822		522

a This has been calculated assuming all of the 300 Kev count in the feed supernate is Pa. This has been found to be nearly true in strong sulfuric acid solution.

The 522 milligrams of protactinium recovered is mostly in a two-molar phosphoric acid solution. Some protactinium has been precipitated from this solution on a ferric phosphate carrier by careful addition of 50 per cent caustic. The development of this process is reported later in this section. It has been found that 70 to 80 per cent of the "iron" in the product phosphoric acid solution is retained in solution using this precipitation method, and nearly 100 per cent of the protactinium is carried by the precipitate, which is readily soluble in dilute hydrochloric acid.

The problem of loss of diisobutyl carbinol (DIBC) in the organic mixture has been lessened by using a mixture of 65 per cent DIBC in Amsco rather than the 50 per cent mixture previously used. Contrary to expectations, the decrease in DIBC concentration seemed to be less using the higher concentration.

Purification - The protactinium concentrate recovered from operations during Fiscal Year 1956 is being homogenized and consolidated preliminary to further purification. It was estimated in last month's report that approximately one month would be required to prepare the necessary homogeneous solution.

Various interruptions, however, including process development, have made it impossible to adhere to this schedule. Approximately one-third of the estimated 700 milligrams of protactinium is now in solution in diisobutyl carbinol. No difficulty has been experienced in preparing this solution, but the operations are being performed slowly and with great caution in order to minimize losses.

The concentration of protactinium in diisobutyl carbinol solution ranges from approximately 0.2 to 2 milligrams per milliliter. Both aqueous and organic waste solutions are being retained and, depending on their concentrations, will be either reprocessed separately or added at appropriate points in the current protactinium recovery process. Before purification operations are started, the solutions containing 0.2 to 1 milligram of protactinium per milliliter will be concentrated further, so that the various solutions can be mixed without excessive dilution of the more concentrated material.

It should be possible to begin purification work by July 15, and it is estimated that the protactinium will be purified and in aqueous solution by September 1.

Process Development - Stripping of protactinium from the organic phase in the first cycle of the protactinium recovery process resulted in large volumes of an aqueous product which was approximately two molar in phosphoric acid. Limited storage facilities made it necessary to reduce the volume of the product, but it was not feasible to carry out the volume reduction by evaporation because of poor ventilation. A precipitate which would carry all of the protactinium and which could be easily separated by decantation was desired.

The following qualitative tests were performed to find the simplest method of forming a small precipitate on which the protactinium could be carried:

1. The solution was saturated with sodium chloride and heated. No precipitate formed except that sodium chloride, which has a very limited solubility in this solution, crystallized out as some of the solution evaporated.

2. Titanium trichloride was added and the mixture was heated. The titanium, which precipitated immediately on addition, dissolved and did not reprecipitate.

3. Sodium hydroxide was added until the solution was alkaline. A very heavy precipitate developed which appeared to be primarily ferric hydroxide. On cooling, the supernate gelled, probably because of the high concentration of sodium phosphate.

4. Sulfuric acid was added, and the solution was heated. No precipitate developed.

5. Calcium hydroxide was added, but produced no precipitate, and only went into solution to a limited extent.

6. Calcium chloride was added. The crystals dissolved and a heavy precipitate formed when the solution was heated. The precipitate, probably calcium phosphate, carried no protactinium.

Although the first calcium chloride experiment was unsuccessful, it seemed possible that calcium chloride could be used as a salting agent for a precipitate with which protactinium would be isomorphous. A number of experiments were performed in which various quantities of calcium chloride and titanium trichloride were added. None of these experiments resulted in quantitative recovery of protactinium. A typical experiment was the following:

> One gram of anhydrous calcium chloride and 10 milligrams of titanium as a 20 per cent solution of titanium trichloride were added to 27 milliliters of the phosphate strip solution. The mixture was heated for one hour at about 90°C, and allowed to cool overnight. The mixture, which had been a clear solution, had a heavy precipitate which was centrifuged off and washed with approximately one-normal hydrochloric acid. The precipitate contained no protactinium. The supernate, reduced in volume to 20 milliliters, was heated with one milliliter of nitric acid (to oxidize titanium), but no precipitate formed after one-half hour at 90°C. The solution was allowed to cool, and an additional 10 milligrams of titanium was added. After one-half hour at 90°C the solution remained clear. Ten milligrams of titanium (for a total of 30 milligrams) was added to the hot solution. Solution of the initial titanium phosphate was incomplete after 15 minutes at 90°C. After one-half hour, a heavy precipitate developed, which had a considerably larger volume than that formed when the titanium was first added. Heating was continued for an additional 15 minutes. The volume had now been reduced to 18 milliliters. Water was added to bring the volume back to the original 27 milliliters, and the solution was allowed to cool to room temperature over a period of several hours. The precipitate was centrifuged, washed with dilute hydrochloric acid, and counted. The precipitate contained 78 per cent of the protactinium.

Numerous other experiments indicated that, while the titanium scavenging of protactinium could probably be made quantitative on a laboratory scale, the operation would not be feasible for large-scale processing.

A series of experiments was next performed to determine if protactinium would be quantitatively carried by a fractional ferric phosphate precipitation. It was found that, if 10-molar sodium hydroxide were added to the phosphate strip solution to the point of incipient turbidity, and if the slightly turbide solution were then heated, a heavy precipitate developed within 10 minutes (at 90°C). The precipitate settled rapidly, and carried protactinium quantitatively.

Because 50 per cent sodium hydroxide is available in the processing area, the necessary conditions were developed for this concentration of alkali. The conditions were found to be moderately critical. When 3.6 milliliters of 50 per cent sodium hydroxide was added to 25 milliliters of phosphate strip solution, no precipitate developed when the solution was heated. When 3.7 milliliters of 50 per cent sodium hydroxide was added to the same volume of strip solution, an excessively heavy precipitate developed on heating. The best ratio for good recovery of protactinium with minimum volume of precipitate was 3.67 milliliters of 50 per cent sodium hydroxide per 25 milliliters of strip solution.

The critical nature of the precipitation was eliminated when the concentration of the added sodium hydroxide was reduced by half. It was therefore, recommended that the following procedure be used:

> To 25 milliliters of phosphate strip solution, add 3.7 milliliters of 50 per cent sodium hydroxide and 3.7 milliliters of water. Mix the solution until only a slight turbidity remains, then heat the solution to about 90°C. Cut off the heat, and allow the mixture to cool while continuing the agitation.

The precipitate settled rapidly on cooling. In less than two hours, the volume of precipitate was approximately one-half the total volume and, on overnight standing, the precipitate settled to one-tenth the total volume, leaving a clear supernate.

A sample of slurry was checked after the precipitation had been carried out on the process scale. It was found that all the protactinium was in the precipitate, and that a significant separation had been

made from other solids in solution. When the supernate was made strongly alkaline with sodium hydroxide and centrifuged, it was found that the volume of hydroxide-precipitable solids in the supernate was approximately 78 per cent of the volume of solids found when the original strip solution was treated in the same way.

Further experiments showed that the ferric phosphate precipitate was completely soluble in dilute hydrochloric acid. When titanium carrier was added to the hydrochloric acid solution of ferric phosphate and the solution heated, the characteristic titanium phosphate precipitate appeared and carried the protactinium quantitatively.

Thus, it appears that it is possible to effect a good volume reduction and separation from iron in two precipitations. The resulting titanium phosphate is readily soluble in sulfuric acid, and can be put through the usual cycle of extraction into diisobutyl carbinol and stripping into phosphoric acid solution. Subsequent separation from titanium is readily accomplished by addition of hydrogen peroxide to the aqueous phase prior to extraction.

Plutonium Urinalysis

Present methods for determining the amounts 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.

No additional work has been done on improving the yield of plutonium in the routine urinalysis program. Because of the limited manpower available, it is not expected that this work will be resumed before October.

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 instituted.

Formulation - The sigma-blade mixer was repaired by replacing the front sigma blade, the drive shaft, and the gear, but a more complete rebuilding will probably be required later as the rear sigma-blade appears to be somewhat out of line, the drive shaft has developed a twist, and difficulties have been experienced with leakage.

Only three formulations were made during this report period because of difficulties with the sigma-blade mixer. The more important data for these runs are summarized in Table I.

TABLE I

Diallyl Phthalate Formulations - Dapon plus Filler, 25 Pounds

	Parts per 100 Parts of Dapon 35							Residual	Peak
Run No.	Filler a/b	Solvent c/d	Monomer	Catalyst e/f	Pigments g/h	Pigments i/j	Zinc Stearate	Volatiles %	Power KW
	40	200		1.75	2.0	0.05	1.5	7.1	2.25
B43			5						
	60	30		0.50	0.2	0.50			
	30	200		1.75	2.2	0.10			
B44			5				1.5	12.3	1.50
	70			0.50	0.1	0.05			
	40	249		2.5					
B45			4				1.5	14.7	1.55
	60	35							

a Cellulo 200
b Hydrite-PD-121
c Acetone
d Benzene
e tert-Butyl Perbenzoate
f di-tert-Butyl Peroxide
g Scarlet 25AD, IR-6440
h Madder Lake R, IR-6050
i Pyrazine Orange ZD-1955
j Pyrazell Red ID-1970

Run B43 was loaded in the sigma-blade mixer in the afternoon, was stirred for two hours, and was then permitted to stand overnight. The next morning, stirring was resumed and, after seven hours total, the mixture began to appear semi-liquid. At the end of the day (with ten hours total mixing time) the mixture, through more liquid and uniform in appearance, was not a homogeneous dispersion. The mixture stood, without stirring from Friday evening to Sunday evening at which time stirring was again started. There was a slow decrease

in input power during each of the three stirring periods. The power was not recorded during the last approximately 16 hours of stirring but it is felt that the power would have decreased negligibly with further mixing.

The acetone was stripped to a 12.4 per cent volatile content, and 30 parts of benzene was added. The mixture was stirred for 30 minutes, then allowed to stand overnight. The solvents were stripped to a volatile content of 7.1 per cent, and the stiff but very plastic-appearing product was removed from the mixer with difficulty. Mixing was unusually good, with partial kneading action obtained throughout the entire run. Kneading action had been practically lost at the time the benzene was added, but was regained shortly thereafter. The last solvent fraction removed had a volume of 340 milliliters of which six milliliters was water. The kneading action appeared to improve as a result of the benzene addition, and a product of lower moisture content should result.

Formulation B44 was run with a 30-to70 ratio of Cellulo-200 to Hydrite-PD-121 to determine processing characteristics and to prepare material for the determination of physical properties. No significant differences were seen between the processing of this formulation and runs using the standard 40-to-60 ratio.

Formulation B45 was run to obtain more data on the use of benzene and to determine the effect of a larger amount of catalyst. The benzene was added (35 parts) after only two liters of acetone had been collected. Kneading action was not nearly as good as was obtained in run B43. Considerable difficulty was experienced with leaking at the shaft seals. This could have been partially responsible for the poor kneading action.

Formulations B42, B43, and B44 were milled on the differential rolls to net power inputs slightly greater than the minimum value obtained after transfer to the hot roll.

Batch B42 had partially polymerized in the sigma-blade mixer, and the unmilled plastic gave a flow time at five tons in the cup flow test of about six seconds. The material was quite stiff and it collected directly on the hot roll. Although very short mill times were used, the products were milled to net power inputs of approximately two kilowatts and hence were quite stiff.

The ground material from the mill runs of B-43 had a strong odor of benzene. It was spread in trays and left on the laboratory bench top over the weekend. The increase in flow time and apparent increase in density, shown in Table II, indicate that drying after grinding is definitely desirable. A sample lost 1.77 per cent of its weight on drying at room temperature in a small vacuum oven over the weekend. This dried sample increased 0.15 per cent in weight on standing on the balance pan. Apparent equilibrium was reached in five hours.

The odor of acetone could be detected in the ground material from the mill runs of B44. Samples were sealed in pint paint cans, while the bulk of the material was dried overnight in trays.

The effect of solvent on flow time was quite marked as shown in Table III. Another difference was in the hardness of the test cups. The cups made from air-dried material averaged 89 on the Rockwell H scale, while the cups from undried material averaged 80.

It had been noticed that there was a variation in the weight of the test flow-cup with the particle size of the material from which the cup was made. The reasons for this variation may be complex, but some variation in residual solvent content with particle size would be expected. The scattered data now available are summarized in Table IV and indicate that residual solvent is a factor.

TABLE II

Effect of Drying on Flow Test Cup Weight - B-43

Sample Number	Flow Time in Seconds at 5 Tons			Weight of Sample Cup in grams		
	5-31-57 at 6 hr	6-31-57 at 72 hr[a]	6-17-57 at 408 hr	5-31-57 at 6 hr	6-3-57 at 72 hr[a]	6-17-57 at 408 hr
B43-1[b]	-	6.65	-	-	39.4	-
B43-1	3.50	6.88	8.62[c]	38.4	39.6	39.6
B43-2	3.14	6.56	8.57	38.4	39.6	39.6
B43-3	3.49	6.55	8.39	38.4	39.5	39.6
B43-4	3.19	6.94	9.04	38.4	39.6	39.6
B43-5	3.43	7.48	9.03	38.4	39.6	39.6
B43-6	3.44	7.22	8.99	38.4	39.5	39.6
Average	3.37	6.90	8.77	38.4	39.54	39.60
Average per cent increase over 6 hr cups					2.97	3.13

a Air dried over weekend spread in trays on bench.

b Dried at room temperature in vacuum oven over weekend; weight loss 1.77 per cent

c Odor of benzene when container was opened for all six samples.

TABLE III

Effect of Residual Solvent on Flow Time - B-44

Net Input Power KW	Sample No.[a]	Flow in Seconds at 5 Tons		Sample No.[b]	Flow in Seconds at 5 Tons	
		at 24 hr	at 144 hr		at 24 hr	at 144 hr
1.34	B44-1	8.36	9.51	B44-1A	6.65[c]	-
1.07	B44-2	6.76	9.04	B44-2A	3.92	-
1.30	B44-3	6.66	8.30	B44-3A	3.49	-
1.16	B44-4	6.49	8.42	B44-4A	3.25	-
1.16	B44-5	6.85	9.21	B44-5A	3.50	-
1.17	B44-6	5.61	8.39	B44-(1+3)A	-	6.11
-	B44-2+5/6	-	9.09	B44(2+5/6)A	-	6.31

a air dried before test

b not dried before test

c Sample B44-1 is believed to have become partially air dried before grinding.

TABLE IV

Variation in Flow Test Cup Weight with Particle Size

Mesh Range	Weight of -16 + 20 Mesh Reference Cup (g) and Sample				
	37.6 MSL-GS	38.67 B35-2/5S	39.4496 B42-1/5S	39.4 B41-1/6S[a]	39.887 B41-1/6S[b]
	Deviation from Reference Cup Weight (-16+20) in Per Cent				
+16	0	-0.44	0.21	-0.51	-
-16 + 20	0	0.00	0.00	0.00	0.00[c]
-20 + 40	0	0.34	0.23	0.33	0.33
-40 + 60	0	1.89	1.06	0.94	0.39
-60 + 80	0	2.15	1.53	1.27	0.68
-80 + 100	0	2.66	-	1.52	-
-100 + 120	0	2.92	-	-	-
-120 + 140	0	3.18	-	2.04	-
-140 + 200	0.27	-	1.82	-	-
-200 + 325	-	-	-	-	-
-200	1.06	3.70	-	2.28	-
-325	-	-	1.99	-	1.72

a Cups molded 5-17-57

b cups molded 6-17-57, reference cup 1.24% heavier

c The average weight of cups molded 6-17-57 of unscreened B41-1 through B41-6 was 40.118 g or 0.58% heavier than reference cups molded from B41-1/6S (-16 + 20 mesh) molded on the same date.

Additional data are being accumulated on the variation of flow time with the age and particle size of the sample, but presentation of these data will be deferred to a later report to avoid excessive repetition of tabular data.

The design of a thermocouple holder for the differential roll has been approved, and one will be constructed for trial. Two millivoltmeters have been ordered for use with the roll-mill thermocouples. The combination should give more reproducible temperature measurement, and should simplify the problem of achieving one-man operation of the differential rolls. It will also be advantageous from a safety standpoint.

Evaluation - Processing of a work order for the procurement of a 40-ton clamp, 10-ton transfer, transfer molding press has been initiated because present molding facilities are inadequate for the preparation of satisfactory test specimens.

Additional test specimens have been molded and, although the quality of most of the specimens was questionable, physical testing was initiated. The data obtained to date are summarized in Table V.

Cure times obtained on heads molded from our materials by Production Molding have generally been low, and it has not been possible to correlate cure times with variation in formulation or flow time. The rather fragmentary evaluation data on heads molded from our materials are summarized in Table VI.

TABLE V

Physical Properties of DAP Formulations

Sample Number	Parts of Monomer	Remarks on Tensile Measurement Breaks	Tensile Strength lb/in²	Arc Resistance Seconds
B34-1/6S1	3	———	—	133.1
B35-6	5	one bad, two good	5,450[a]	-
B37-1/6F1	5	two good	5,650	-
B37-1/6F2	5	two good	3,910[b]	-
B40-1/6	5	one each good, fair, bad[c]	5,948	135.3
B41-1/6S2	5	one good, one bad[c]	5,867	140.0
B41-1/6S3	5	one good, one bad	5,464[d]	148.0
Mesa B 1143	?	three good	5,476[e]	131.6

a Average of two good breaks
b Value probably not representative
c Bad Breaks gave highest value
d Bad break 4,603 psi, good break 6,325 psi
e A value of 5,630 psi was obtained in previous measurement.

TABLE VI

Evaluation of Heads Molded from Mound Laboratory DAP

Sample Number[a]	Number of Heads Molded	X-ray Inspection Data Cracks %	Voids %	Inclusions %	Cure Time Min.	Date Heads Molded
B14	15	100	-	-	1.50	12-11-56
B16	15		-	-	3.30	1-4-57
B30-6	100	72.6	27.4	-	2.30	3-11-57
B34	97	85.9	8.7	1.1	1.50	3-29-57
B36-1/6S1	100	77.0	8.4	3.2	1.70	4-4-57
B35-6	50	51.0	6.7	4.1	1.30	4-30-57
B35-2/5-S1	35	100.0	-	-	2.04	5-3-57
B35-2/5-S2	35	97.0	-	-	1.44	5-3-57
B35-2/5-S3	35	100.0	-	-	0.99	5-3-57
B37-1/6C	275	20.8	2.0	16.3	2.00	5-11-57
B41-1/6S3	208	-	-	100.0[b]	1.20	5-22-57

a B30-6: 2% catalyst, 2% monomer
B34: 2% catalyst, 3% monomer, 1.48 KW net
B35-1/6: 2% catalyst, 3% monomer, 1.32 KW net
-S1: -8 + 16, S2: -16 + 20, S3: -20 + 40 mesh
B36-1/6S1: 2% catalyst, 5% monomer, -8 + 16 mesh
B37-1/6C: 2% catalyst, 5% monomer, -100 mesh removed.
B41-1/6S3: 2.25% catalyst, 5% monomer, 1.16 KW net, -20 + 40 mesh.

b Cellulo 200 used contained magnetite.

Heads molded from Durez DAP have usually given low values for cure time though no significant change in appearance could be noted as a result of the extraction. Four old, rejected heads, each of a different color, and one of the early heads molded from Mound DAP were chosen at random. The hardness was determined for each, they were chloroform extracted, and the hardness was again determined. The data are summarized in Table VII.

TABLE VII

Comparison of Five Formulations on Cure and Hardness

Formulation Assumed from Color of Head	$CHCl_3$ Extraction Cure		Rockwell H Hardness	
	Δ Thickness min.	Δ Diameter min.	Before Extraction	After Extraction
Mesa Natural	9.7	19	92	90
Acme Blue	1.43	1.29	88.5	90
Acme Natural	1.08	0.90	86	86
Durez Green	0.95	0.78	91	92
Mound White	0.58	0.46	83	78

A series of flow test cups were made with mold closure times varying from 0.5 to 20 minutes for one batch of Mesa Natural, and for two Mound formulations. The hardness increased rapidly at first but soon reached a nearly constant value. The results to date are summarized in Table VIII.

It would appear that the hardness test will readily detect badly under-cured moldings, but much more extensive work will be required to determine if hardness can be used as an acceptance test. Picatinny Arsenal has been doing some work on hardness as a measure of cure. They have used the Rockwell M scale (¼-inch ball, 100 kilogram load). Their tentative conclusion is that hardness may not be a sufficiently sensitive method for use in determining cure above some not yet established medium value.

Production Service - A complete flow curve was run on Mesa Natural DAP batch B-1433 which had been ordered with a flow specification of 15 ± 1 seconds at 5 tons. A flow time of 15.16 seconds at 5 tons was measured.

A tentative equipment list and flow chart for the Plastic Production Facility have been worked out. Inquiries have been sent to several equipment manufacturers to assist in the determination of equipment specifications for the Plastic Production Facility.

Dry-blending experiments were run at the J. H. Day Company, Inc., on DAP molding powder and on some of our dry ingredients. A test of screen capacity for DAP molding powder was also made. Evaluation is incomplete. The ribbon-type blender appears to be satisfactory for blending DAP molding compounds. The operation was very dusty, and the power requirements were high. Kaolinite had a tendency to form balls in dry blending, and this may be a problem. There appear to be no serious problems in screening DAP molding compounds, but the operation is very dusty.

TABLE VIII

Variation of Hardness of Cups with Mold Closure Time

Mold Closure Time min.	B34-1/6S1 Flow at 5 Tons, sec	B34-1/6S1 Hardness Rockwell H Scale[a]	B39-2 Flow at 5 Tons, sec	B39-2 Hardness Rockwell H Scale	Mesa B1433 Flow at 5 Tons, sec	Mesa B1433 Hardness Rockwell H Scale
0.5	11.10	b	11.11	36	17.68	b
1.0	12.37	b	11.83	72.5[c]	18.06	b
1.5	10.87	81	10.94	71	17.06	84.0
2.0	11.47	88	10.50	88.9	16.45	87.0
2.5	10.66	87.0	11.27	90.7	16.80	88.8
3.0	10.40	91.8	11.31	93.8	17.15	92.2
3.5	10.47	93.8	10.74	94.7	16.17	94.2
4.0	10.31	93.8	10.90	95.7	15.96	94.2
5.0	11.10	94.4	10.75	96.9	15.66	92.8
6.0	10.64	94.8	10.69	97.0	16.55	95.4
7.0	11.05	96.4	10.94	96.6	16.76	94.5
9.0	10.56	94.2	10.71	96.0	16.41	94.2
12.0	11.14	96.8	10.10	96.8	16.94	94.2
15.0	10.66	95.4	10.94	97.3	16.70	95.4
20.0	11.72	96.4	10.99	97.2	16.86	95.0
60.0	-	-	11.14	98.0	-	-

a 1/8 inch ball, 60 kilogram load

b Sample cracked under load

c Where hardness is given to three significant figures, value is the average of five or more measurements. It should be noted that reproducibility does not warrant conclusions, based on three significant figures.

Very long mixing times in the sigma-blade mixer are required, and it is felt that defelting is incomplete even then. It would therefore be highly advantageous if the mix could be made more fluid and could be more completely defelted before being added to the sigma-blade mixer. It is believed that high-speed stirring, giving high localized sheaf forces, would give better defelting if the mixture could first be made fluid to a point where high speed stirring could be used.

A series of stirring experiments were run at Chemineer, Inc. in Dayton, Ohio. Several types of standard impellers were tried, both singly and in combination, but in no case could a major portion of the material be moved. Special stirring or mixing equipment will apparently be required. The Patterson Foundry and Machine Company has been contacted, and tests will probably be run at their plant in East Liverpool, Ohio.

Dry-blending tests have been made by Patterson-Kelley Company, Inc. The laboratory report has not been received. The samples have arrived at Mound Laboratory but have not been evaluated.

The last 300-pound order of Cellulo-200 asbestos was found to contain a small amount of magnetite. Most of this was present as relatively large particles, hence X-ray rejects for inclusions were very high. Mr. Knauer of the Cellulo Company was contacted by telephone. He said that he had had difficulty with his supplier. He requested that we return this material, and that he would replace it at no cost to us.

This incident indicates the desirability of establishing acceptance tests for Cellulo asbestos, and that material control should probably be established for all of our raw materials.

GENERAL WEAPONS DEVELOPMENT

Process development work is being conducted on the use of thermal diffusion for the separation 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 - Continuous-flow separation of oxygen isotopes with the four 12 foot-long columns in a series cascade yielded a gas containing 4.4 per cent oxygen-18 and 0.18 per cent oxygen-17. A sample was obtained after approximately one week's operation. The normal oxygen was fed at a point six feet below the top end of the cascade; the four Nichrome wires were maintained at 900°C. The operating pressure of the cascade was 180 millimeters of mercury; the cold pressure was 130 millimeters of mercury.

The above concentration of oxygen isotopes seems to represent the equilibrium separation of the cascade under these conditions. Since major interest is in enriched oxygen containing at least 50 per cent oxygen-17 plus oxygen-18 (which could be obtained only after several months of single, closed-end operations with this facility), work on this separation problem has been discontinued for the time being.

Work was started on the separation of neon isotopes in this cascade system using a neon feed of normal abundance. After one week's operation at a pressure of 350 millimeters of mercury and with center-wire temperatures of 900°C, samples containing approximately 40 per cent neon-22 were obtained. As this concentration was considerably below expectations, a mass scan was made for contaminants in the samples. This scan revealed the presence of considerable quantities of masses 28 and 32, indicating contamination by nitrogen and oxygen from air leakage into the system. The cascade system is now being leak-tested and repaired. Neon-22 enrichment will then be continued.

The gas-standard preparation system has been reassembled, and a series of gas-standard samples will be prepared for recalibration of the mass spectrometer.

A total of 28 oxygen samples and 27 neon samples were analyzed on the mass spectrometer this month. This work was done in conjunction with the above experimentation. The mass spectrometer was shut down for two days this month for routine maintenance.

Gas Purification - Leak testing of the entire gas purification system under 50 psig helium pressure with the helium leak detector was completed. Leaks were found and eliminated in the Poro-stone filter, in the flange on the cold trap, and in several valves. The cold trap was tested for leakage while filled with liquid nitrogen, and was found to be satisfactory.

All the Teflon diaphragms in the Red Head valves were cleaned after being found to have accumulated dirt and dust on their sealing surfaces.

A final check showed that it was possible to evacuate the entire system to less than one micron pressure. The uranium pumps were outgassed to that pressure in preparation for activation with hydrogen.

Liquid Thermal Diffusion - The study of the effect of time on the per cent separation for different pairs of fractions has been extended with a two-hour run using the benzene-carbon tetrachloride system. As before, the hot and cold walls were held at nominal temperatures of 50 and 17°C, respectively. The per cent separation obtained, based on refractive index measurements, beginning with that based on the end fractions and receding progressively from the ends was 56.5, 28.2, 15.6, and 8.4 per cent. These values are consistent with those reported last month for runs extending from 4 hours to 48 hours.

The effect of temperature gradient and wall temperature on separation has been studied; the results are reported in Table I. Increases in hot wall temperatures with consequent increases in gradient effect marked increases in separation It should be noted that the results are based on 6-hour runs and do not represent equilibrium or steady-state conditions. The last run reported in the table was obtained with the heater on full power. The measured voltage drop through the heater was 108 volts and the measured current was 17.7 ampers.

TABLE I

Effect of Wall Temperatures and Temperature Gradient on Per Cent Separation for Different Pairs of Fractions

(Feed: 50-50 Volume per cent C_6H_6-CCl_4. Time 6 hours)

Av. Hot Wall °C	Av. Cold Wall °C	Mean °C	Difference °C	Per Cent Separation Based on Fractions 1 and 9 (end)	2 and 8	3 and 7	4 and 6
30.3	15.7	23.0	14.6	31.8	12.3	6.4	3.7
50.0	17.0	46.5	33.0	87.5	81.0	58.5	31.0
64.3	17.7	55.3	46.6	96.8	96.4	92.1	76.2

Preliminary tests with the annulus filled with ordinary distilled water have revealed several significant differences in behavior as compared with the organic feed. In tests with water, little or no gas was observed to accumulate at the top of the annulus even when the column was filled without prior evacuation. Also, the temperature gradient obtainable between walls with the annulus filled with water was less than that obtained with the organic feed, presumably because of the higher thermal conductivity of water. Thus, with the heater operating at full power and with a cooling water flow rate of four liters per minute, the C_6H_6-CCl_4 system gave a temperature difference of 46.5°C between walls whereas natural water, as well as a 50-50 volume per cent mixture of heavy water and ordinary water, gave a temperature difference of approximately 15°C.

The possibility of increasing the temperature gradient between walls by reducing the cooling water flow rate was checked. Table II shows the dependence of wall temperatures on cooling water rate when the annulus is filled with ordinary water. The data show that decreasing the cooling water flow rate will increase the average gradient between walls but only at the expense of increasing the vertical gradients along the walls. The use of refrigerated cooling water as a means of increasing the gradient is under consideration.

TABLE II

Effect of Cooling Water Flow Rate on Wall Temperatures

(Annulus filled with distilled water. Amperage = 17 (Maximum)

Water Rate (liters per minute)	Hot Wall Temperatures, °C					Cold Wall Temperatures, °C				Average Difference Between Walls °C
	Bottom	Middle	Top	Ave.	Range	Inlet	Outlet	Ave.	Difference	
1.89	35.0	43.5	50.5	43.0	15.5	16.0	26.0	21.0	10.0	22.0
4.00	32.5	34.5	39.0	35.3	6.5	16.0	20.5	18.3	4.5	17.0
7.20	32.0	30.5	35.0	32.5	4.5	16.0	18.5	17.3	2.5	15.2

A 48-hour run was completed using a 50-50 volume per cent mixture of H_2O and D_2O as feed. The column was operated at full power, giving a hot wall temperature of 35°C, a cold wall temperature of 20°C, a temperature difference of 15°C. The fractions will be analyzed by specific gravity measurements. The installation of the second liquid thermal diffusion column has been completed. The first three points of the six-point recorder are being used to record wall temperatures of the first column and the last three points to record the wall temperatures of the second column. By means of a series of double-throw, double-pole switches, all six wall temperatures may be recorded, though not all simultaneously

Several determinations of the annulus width of the second column indicate a value of 0.0125 inches. Simultaneous operation of both columns charged with distilled water indicates that their thermal characteristics are similar

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 made on an aqueous system containing up to 300 grams of uranium per liter and five grams of plutonium per liter.

Five series of runs were made on the solubility of the system plutonium (III) sulfate-sulfuric acid-water. The results of three of these series are listed in Table I. The plutonium (III) in one series of solutions was found to have been partially oxidized to plutonium (IV) in solution, and this series is not listed. Investigation showed that this oxidation could be prevented by displacing some of the air over the solution with sulfur dioxide. The fifth series gave measurements inconsistent with previous results and is being repeated.

TABLE I

Plutonium (III) Sulfate Solubility in Sulfuric Acid-Water Solutions

0.067 N H_2SO_4		0.10 N H_2SO_4		0.6 N H_2SO_4	
Temp. of Phase Change °C	Pu Conc. g/liter	Temp. of Phase Change °C	Pu Conc g/liter	Temp of Phase Change °C	Pu Conc. g/liter
166.2	13.28	167.2	18.71	202.0	19.52
174.8	9.49	172.0	12.00	191.8	13.53
197.2	5.69	182.2	8.55	224.4	12.00
204.8	3.79	205.0	6.85	242.2	7.23
233.2	1.90	201.2	4.14	335.5	1.90[b]
249.0	1.14	252.8	1.91[a]	338.0	1.33[c]
262.0	0.76				

a Two liq. phases, 215°C
b Two liq. phases, 235°C
c Two liq. phases, 220°C

One dilute sample in the 0.10 N H_2SO_4 series and two dilute samples in the 0.6 N H_2SO_4 series showed formation of a second liquid phase at about 220°C. This phase appeared as brown droplets which coalesced into a tiny bead and became smaller and darker as the temperature was raised. On cooling, the beads again grew to original size, but did not form a single-phase liquid again. At room temperature, the drop became a yellow, waxy solid. Investigation showed the material to be elemental sulfur. No explanation has been found for the sulfur reduction. Hyposulfite formation from the sulfur dioxide atmosphere seems very unlikely in view of the pH of the solution and the large amounts of sulfur formed. The plutonium remained in the plus three oxidation state, as evidenced by the blue color of solution, and the quartz tubes remained clear and unchanged.

A method for preparing and evaluating plutonium (III) carbonate solutions has been devised for use with the quartz microtubes. It is presently being checked for accuracy using lanthanum as a substitute for plutonium. In brief, the plutonium is precipitated as the hydroxide in the quartz tube and centrifuged. After washing with water and recentrifuging, water and carbon dioxide are added. The carbon dioxide is condensed by freezing it (as well as the solution) in the quartz tubes with liquid nitrogen and kept frozen while the tube is sealed. The fluids are again frozen before opening the tubes for analysis. Analysis involves determining water content from the weight of solution, alpha counting for plutonium assay, and collecting and weighing the carbon dioxide in a small weighing bulb containing Ascarite.

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 - Liquidus temperatures obtained to date from the differential thermal analysis studies of different mixtures of LiF, RbF, and BeF_2 are summarized on Figure 1. These data include thirty-seven values determined since the last report.

The phase equilibrium and temperature contour diagram showing the interpretation of data obtained up to the present time appears as Figure 2. The invariant equilibria points assigned at this time are designated by capital letters. These points are listed in Table I. This diagram was constructed on the basis of differential thermal analysis data from 115 different compositions, the identification of primary phases and boundary intersections of thermal gradient quench samples and slowly-cooled compositions, high temperature filtration studies, and X-ray diffraction data. Recent results of thermal gradient studies of various ternary compositions involving identification of primary phases and boundary intersections are listed in Table II. Results of high temperature filtration studies are shown in Table III. The composition triangles, indicated by dotted lines on Figure 2, were constructed on the basis of the locations of eutectic, pertiectics, and the two ternary compounds which appear to be in this system.

The phase boundaries between the BeF_2 and the $RbBe_2F_5$ primary phase area, and between the "$Li_2RbBe_2F_7$" and the $RbBeF_2$ primary phase areas and the location of peritectic "B", are designated by dashed lines to indicate that some uncertainty exists in this portion of the phase equilibrium diagram. In the BeF_2-rich areas the thermal effects of the transitions are small and thus poorly defined in the differential thermal analyses studies. Only the liquidus valves are considered to be reliable. The location of the point "B" is proposed to satisfy the requirements of the established composition triangles. Limited information based on the identification of the primary phases from thermal gradient-quenched samples suggests that the primary phase area for the ternary compound, tentatively identified as $Li_2RbBe_2F_7$, should be changed toward a greater BeF_2 content than is currently indicated on Figure 2. Composition R-21, Table I, examined after thermal gradient quenching, had "$Li_2RbBe_2F_7$" as a primary phase at an unusually low liquidus temperature of 310°C. This low temperature may have resulted from supercooling. An increase in the size of the primary phase area assigned to "$Li_2RbBe_2F_7$" would mean a decrease in the size of the $RbBe_2F_5$ primary phase area. Also, it should be noted that $RbBe_2F_5$ has not been observed as a primary phase in quenched samples that contained less than 22 mole per cent RbF.

[redacted]

LIQUIDUS TEMPERATURES (°C)
OBTAINED BY D.T.A. FOR
THE SYSTEM LiF–RbF–BeF_2

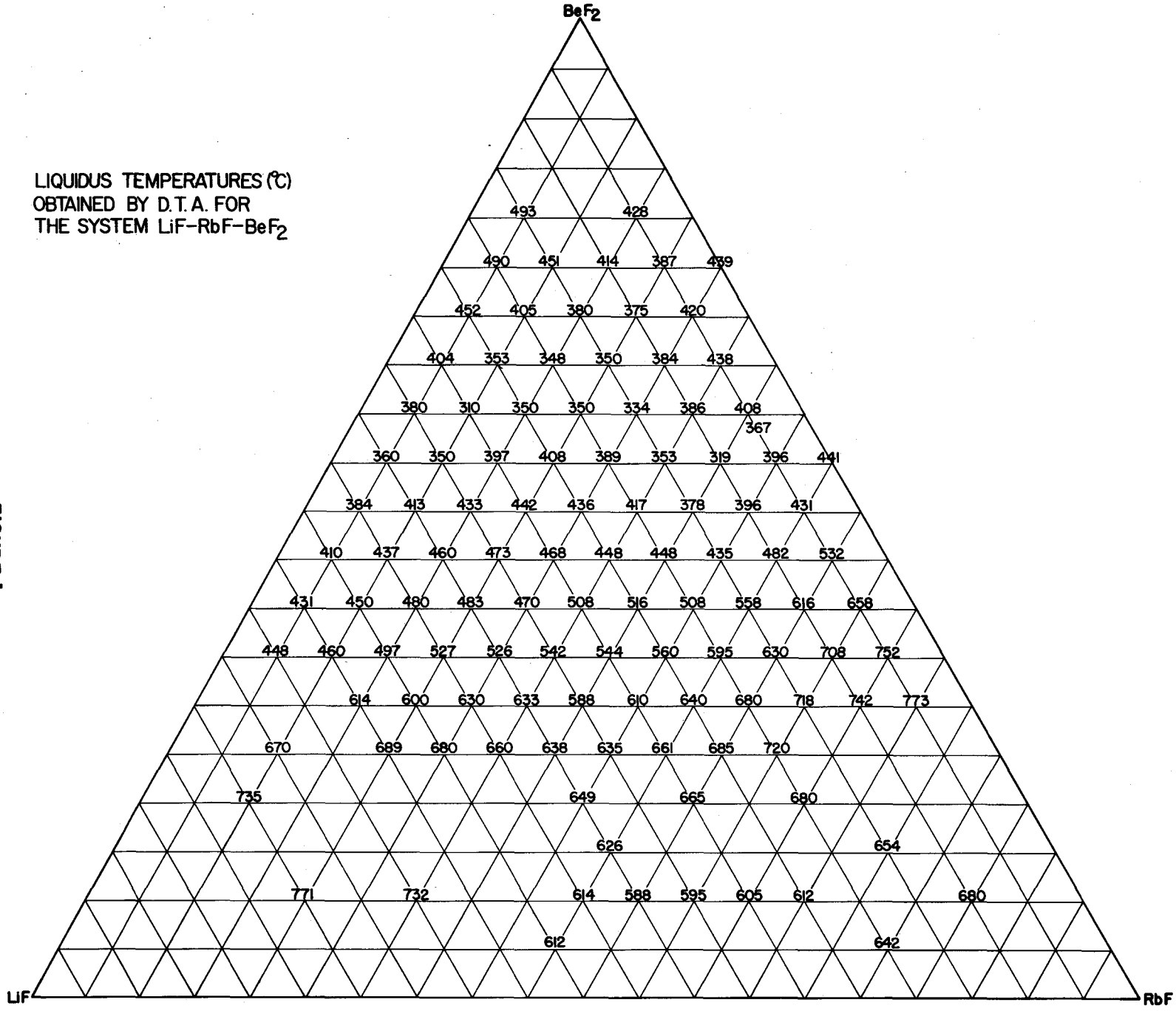

FIGURE 1

[redacted]

~~SECRET~~

PHASE DIAGRAM AND TEMPERATURE CONTOURS FOR THE SYSTEM $LiF-RbF-BeF_2$

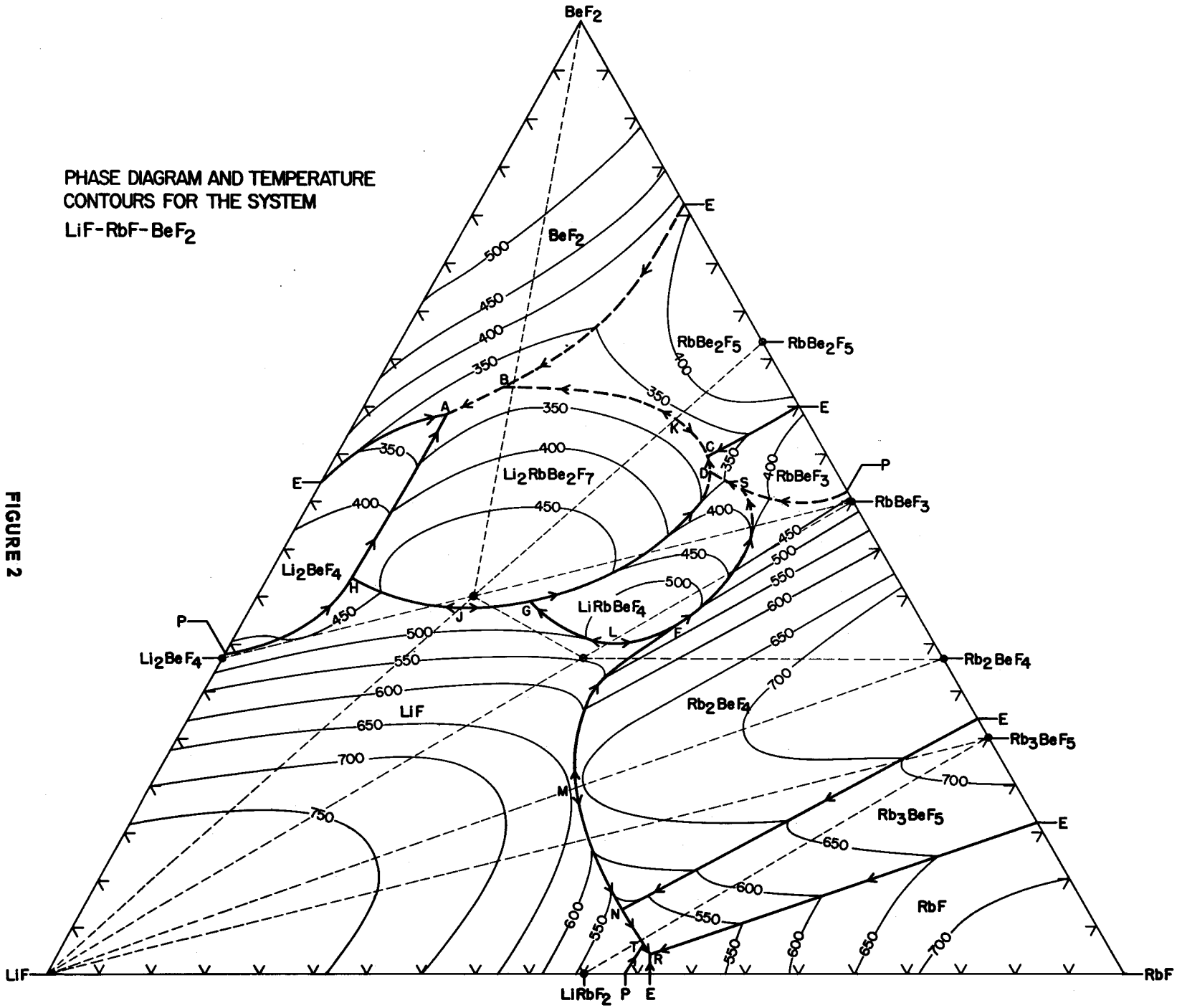

FIGURE 2

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TABLE I

Invariant Equilibria Shown on Figure 2

Location	Phase	Type	Temperature °C
A	$Li_2RbBe_2F_7 \cdot Li_2BeF_4 \cdot BeF_2$	Ternary Eutectic	295
B	$Li_2RbBe_2F_7 \cdot RbBe_2F_5 \cdot BeF_2$	Ternary Peritectic	?
C	$Li_2RbBe_2F_7 \cdot RbBe_2F_5 \cdot RbBeF_3$	Ternary Eutectic	300
D	$Li_2RbBe_2F_7 \cdot LiRbBeF_4 \cdot RbBeF_3$	Ternary Peritectic	335
F	$LiRbBeF_4 \cdot LiF \cdot Rb_2BeF_4$	Ternary Peritectic	530
G	$Li_2RbBe_2F_7 \cdot LiF \cdot LiRbBeF_4$	Ternary Peritectic	470
H	$Li_2RbBe_2F_7 \cdot LiF \cdot Li_2BeF_4$	Ternary Peritectic	426
J	$Li_2RbBe_2F_7 \cdot LiF$	Quasi-binary Eutectic	473
K	$Li_2RbBe_2F_7 \cdot RbBe_2F_5$	Quasi-binary Eutectic	334
L	$LiRbBeF_4 \cdot LiF$	Maximum on Boundary	544
M	$LiF \cdot Rb_2BeF_4$	Quasi-binary Eutectic	640
N	$LiF \cdot Rb_2BeF_4 \cdot Rb_3BeF_5$	Ternary Peritectic	527
R	$LiRbF_2 \cdot Rb_3BeF_5 \cdot RbF$	Ternary Eutectic	462
S	$LiRbBeF_4 \cdot RbBeF_3 \cdot Rb_2BeF_4$	Ternary Peritectic	380
T	$LiRbF_2 \cdot Rb_3BeF_5 \cdot LiF$	Ternary Peritectic	?

The ternary compound tentatively identified as $Li_2RbBe_2F_7$ melts congruently between 478°C and 483°C and appears to have a relatively large primary phase area. Differential thermal analyses of ternary compositions on and just to each side of the join between LiF and "$Li_2RbBe_2F_7$" established this join as a quasi-binary, and the invariant point, J on Figure 2, as a quasi-binary eutectic. Similar studies showed that the join between "$Li_2RbBe_2F_7$" and $RbBe_2F_5$ is a quasi-binary with its eutectic at the invariant point, K on Figure 2. Differential thermal analyses of ternary compositions on and to each side of the join between LiF and a second ternary compound, identified as $LiRbBeF_4$, indicated that a maximum invariant point, L on Figure 2, exists on the LiF-$LiRbBeF_4$ phase boundary. The direction of the crystallization path of all compositions in the primary phase area assigned to $LiRbBeF_4$ were away from the phase boundary, indicating that this compound melts incongruently.

Single crystals were separated from a thermal gradient quench sample which contained an equimolar composition of LiF, RbF, and BeF_2. One of these crystals which showed a good extinction was mounted and examined by single-crystal X-ray techniques. Precession and Weissenberg photographs were made from which the space group and unit cell were determined. The crystal was hexagonal. All hkil were present, 0001 being present when l = 2n. It was found that a = 5.201 ± 0.005 Å, c = 8.761 ± 0.005 Å, V = 205.2 Å^3. Possible space groups indicated by the extinctions are D_6^6 – $P6_322$, C_6^6 – P_3, or C_{6h}^3 – $P6_3/m$.

TABLE II

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

Comp. No	Composition, Mole Per Cent LiF	RbF	BeF_2	Phase Change Temperature °C	Phases Found Just Above Phase Change	Phases Found Just Below Phase Change	Interpretation
R-21	17	18	65	310	Liquid	Liquid, Comp. A	Liquidus and primary phase
R-27	30	20	50	450	Liquid	Liquid, Comp. A	Liquidus and primary phase
R-31	10	45	45	465	Liquid	Liquid, Rb_2BeF_4	Liquidus and primary phase
R-33	30	25	45	470	Liquid	Liquid, Comp. A	Liquidus and primary phase
R-34	40	40	20	655	Liquid	Liquid, Rb_2BeF_4	Liquidus and primary phase
	40	40	20	645	Liquid, Rb_2BeF_4	Liquid, Rb_2BeF_4, LiF	Boundary intersection
R-35	53	6	41	445	Liquid	Liquid, Li_2BeF_4	Liquidus and primary phase
	53	6	41	413	Liquid, Li_2BeF_4	Liquid, Li_2BeF_4, Comp A	Boundary intersection
R-36	38	27	35	536	Liquid	Liquid, LiF	Liquidus and primary phase
	38	27	35	–	Liquid, LiF	Liquid, LiF, Comp. B	Boundary intersection
R-38	23	42	35	580	Liquid	Liquid, Rb_2BeF_4	Liquidus and primary phase
	23	42	35	545	Liquid, Rb_2BeF_4	Liquid, Rb_2BeF_4, Comp B	Boundary intersection
R-39	40	20	40	478	Liquid	Liquid, Comp. A	Liquidus and primary phase

TABLE III

Liquidus Values by Filtration

Comp. No.	Starting Composition Mole Per Cent LiF	RbF	BeF_2	Filtration Temperature °C	Filtrate Composition Mole Per Cent LiF	RbF	BeF_2	Interpretation of Location of Filtrate on Phase Diagram
1	40	30	30	540	34.7	33.4	31.9	Maximum on boundary LiF-$LiRbBeF_4$
2	20	40	40	390	13.9	39.9	46.2	Possible boundary between $LiRbBeF_4$-Rb_2BeF_4
3	50	10	40	406	49.8	8.1	42.1	Boundary, $Li_2RbBe_2F_7$-Li_2BeF_4
4	65	5	30	431	59.5	6.7	33.8	On LiF phase area
5	55	15	30	470	45.3	17.7	37.0	On LiF phase area
6	15	70	15	550	36.9	58.8	4.3	Boundary, RbF-Rb_3BeF_5
7	15	70	15	560	31.0	61.7	7.3	Boundary, RbF-Rb_3BeF_5

The density of the single crystal was measured by suspending it in a liquid whose specific gravity was adjusted to be just equal to that of the crystal. The density of the suspending liquid was determined to be 2.92 g/cm^3 by displacement of a calibrated platinum sinker.

The formula weight of the substance was calculated from its density and unit cell dimensions to be 363.1/m, where m equals the number of molecules per unit cell. The symmetry of the space groups allows m to be 2, 4, 6, and 12. Therefore, the weight of the molecule can only be 181.6, 90.78 or 60.52. $LiRbBeF_4$, whose molecular weight is 177.40, is the only compound in this ternary system which gives a reasonable fit. Back calculations indicate the density by X-ray is 2.85 g/cm^3. Agreement between the calculated and experimentally-determined densities is about 2.4 per cent. The formula of this ternary compound is $LiRbBeF_4$. This formula eliminates C_6^6 -$P6_3$ as a possible space group since there are no four-fold positions in this space group. Therefore, the space group is either D_6^6 -$P6_322$ or C_{6h}^2 -$P6_3/m$.

DEPARTMENT OF DEFENSE

U. S. ARMY SIGNAL ENGINEERING LABORATORIES

Ft. Monmouth, N. J.

COMPONENTS BRANCH

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

Nuclear Battery - Thermoelectric Type

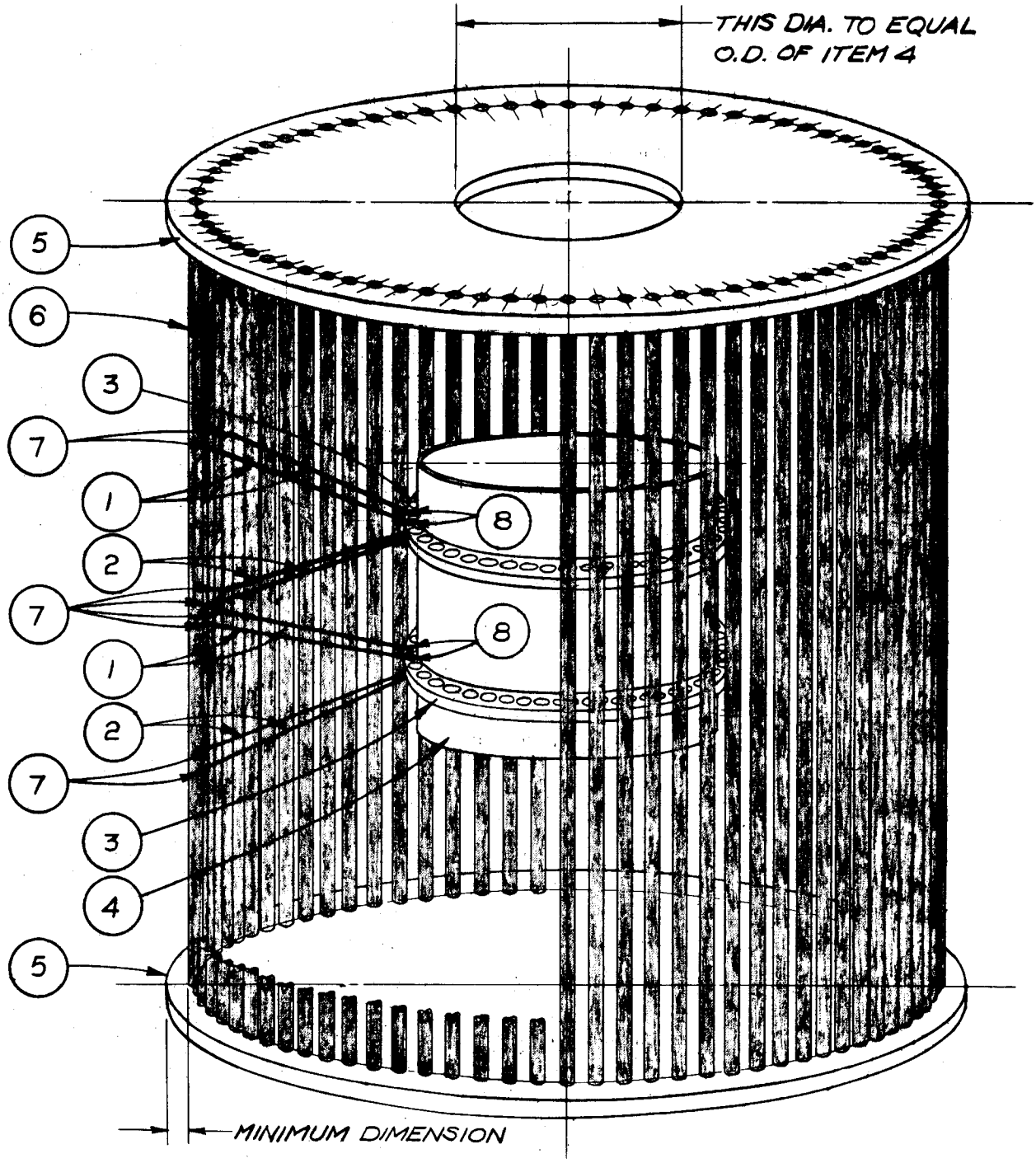
The development of a system for the direct conversion of the energy of radioactive decay as heat into electrical energy is under investigation for the U.S. Army Signal Engineering Laboratories by Mound Laboratory. Radioactive materials are being evaluated as heat sources with consideration given to availability,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

Work was begun on a prototype thermoelectric generator in order to test materials and techniques of construction. The generator, shown in Figure 1, contains 120 Chromel-Constantan junction pairs and should develop about two volts for a temperature difference of 400°C.

The two materials tested for the heat source container were nickel and aluminum. It was hoped that a sufficiently heavy anodized coat could be applied so that bare thermocouple wires could be threaded through the holes and still remain electrically insulated from the source. Attempts to anodize the aluminum in sulfuric acid and in chromic acid baths as well as in commercial bath solutions gave coats which were unsatisfactory for this purpose. However, anodizing 2S aluminum in seven per cent sulfuric acid at 18 volts for three hours produced a coat which may be satisfactory.

Tests indicated that a very tight, superior coat was produced on nickel when oxidized above 900°C. The electrical sensitivity of this coat falls from greater than 10 megohms resistance per mill at room temperature to several thousand ohms at 600°C. This coat is very hard and dense, and is quite resistant to abrasion.

The thermocouples are made of No. 28 Chromel and No. 30 Constantan wire spot welded together at the hot junction. At the cold junctions they are silver-soldered to No. 20 Formax-insulated copper wire. The copper wires form a cage to retain insulating material within a top and bottom support plate made of 1/16-inch thick Formica.



1 - Type 1 Wire
2 - Type 2 Wire
3 - Equatorial Rings
4 - Source Container
5 - Supporting Insulating Plates
6 - Conducting Support Rods
7 - Cold Junctions
8 - Hot Junctions

FIGURE 1