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MONTHLY TECHNICAL ACTIVITIES REPORT THROUGH JANUARY 15, 1958

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MOUND LABORATORY
Miamisburg, Ohio
Operated By
MONSANTO CHEMICAL COMPANY
Research and Engineering Division

United States Atomic Energy Commission
Contract Number AT-33-1-GEN-53

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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.

Seven polonium-beryllium neutron sources and five alpha sources were shipped in December, 1957. Five polonium-beryllium sources were for other AEC sites. There were twenty-two plutonium-beryllium neutron sources shipped during December, 1957.

On January 6, 1958 the conversion factor for curies of polonium used in the calorimeter calculations was changed from International Watts to Absolute Watts. This was done to bring the factor into agreement with the component parts of the calorimeter console which are calibrated in Absolute Watts. The change was from 31.240 to 31.235 and should result in more accuracy, especially on high curie value items.

Twenty-four tanks of influent water were processed during this period. Two of the tanks required no pre-treatment. Thirteen of the tanks required one treatment of activated carbon and calcium chloride. Nine tanks required a second treatment of sodium sulfide and barium chloride. Data for the Waste Disposal Operations are given in Table 1.

Table 1

WASTE DISPOSAL DATA

Discharge Volume	670,600	gallons
Total Alpha Activity	8.8	millicuries
Total Beta Activity	45.6	millicuries
Activity Density		
Alpha	3.8	cts/min/ml
Beta	19.9	cts/min/ml

NUCLEAR RESEARCH AND DEVELOPMENT

GENERAL WEAPONS DEVELOPMENT

Process development work is being conducted in connection with the fabrication and chemical processes for plastics, high explosives, gases, metals, alloys, and other materials for weapons components, and the fabrication and recovery associated therewith.

Plastics Project *The asbestos-filled diallyl phthalate plastic used in molding operations at Mound Laboratory must meet specifications more rigid than are common in industry. Development work has been directed at the determination of the effects of process variables and toward achievement of better control of these variables in order that plastic which meets specifications may be consistently produced. In addition, efforts will be made to find plastics and fillers which would provide improved properties.*

Formulation Only two sigma-blade mixer runs were made during this report period. These formulations are summarized in Table 1.

Table 1

DIALLYL PHTHALATE FORMULATIONS

Run Number	Parts Per 100 Parts Dapon-35						Wt. of Charge Minus Solvent lb	Peak Input Power KW	Residual Volatile Content %
	Filler	Acetone	Monomer	Catalyst	Calcium Stearate	Hydroquinone			
B84	40[a]	235	0.0	2.25	3.0	0.1	27.3	1.30	15.0
B85	100[b]	270	4.0	2.0	2.0	0.0	26.0	1.60[c]	11.5

a 40 Parts Cellulo-200
b 40 Parts Cellulo-200 plus 60 parts Hydrite PD-121
c Peak of 1.6 KW caused by material stuck to the sides being drawn into the mixing zone. Peak power input just prior to this was 1.30 KW.

Mixer run B84 was an experimental run made without kaolinite. The asbestos was prewet with dilute varnish by mixing for two hours in the sigma-blade mixer. The prewet asbestos was then added to filtered varnish in a nine-gallon Pyrex cylinder with stirring. The asbestos suspended and stirred well at first, but the mix became semi-solid before all of the asbestos had been added. Stirring was continued and eventually the entire mass became fluid. High-speed stirring (200-2,000 rpm) was continued for 10 hours, after which the material was transferred to the sigma-blade mixer for removal of acetone.

Three four-inch turbine impellers placed one above the other were used in stirring. The lower turbine was pitched to stir up, the center impeller had flat blades (90° pitch), and the upper turbine was pitched to stir down. This arrangement appeared to give good action, but impellers of larger diameter would permit a greater amount of work to be done on the suspension.

Kaolinite was again used in run B85. The asbestos was prewet with 60 parts of dilute varnish in the sigma-blade mixer, and the kaolinite was prewet with 40 parts of varnish in a beaker. The prewet asbestos was added to the varnish at a slower rate than for run B84. Complete stirring was obtained more rapidly as a result. The slurried kaolinite was added after eight hours of high-speed stirring, and stirring was continued for 16 additional hours. This resulted in a very smooth and creamy mixture of good fluidity which appeared to be very homogeneous.

The compositions after milling for the 35 lots processed on the differential rolls are given in Table 2. The milling conditions and cup flow test times are given in Table 3.

Runs B73, B74, and B80 were designed primarily to obtain information on the effects of mill loading, roll spacing, roll temperature, and input power on the amount of air entrained in the plastic during milling. The other runs were primarily directed at determining the optimum concentration for monomer, catalyst, and inhibitor.

Table 2

COMPOSITION OF DAP FORMULATIONS AFTER MILLING

Mill Lot	Parts per 100 Parts of Dapon-35								
	Filler			Stearate			Catalyst		$CHCl_3$
No.	Cellulo	Hydrite	Monomer	Calcium	Zinc	Hydroquinone	TBP	DTBP	Cure min
B73-3	40	60	5	3	—	—	2.5	—	11.2
B73-4 AD	40	60	5	3	—	—	2.5	—	20.1
B73-5 AD	40	60	5	3	—	—	2.5	—	7.6
B73-6 AD	40	60	5	3	—	—	2.5	—	—
B74-4	40	60	5	2	2	0.02	2.75	—	3.8
B75-5	40	60	5	3	1	—	2.5	—	—
B75-6	40	60	5	3	1	1.00	2.5	—	No cure
B76-3	40	60	5	3	1	0.10	2.5	—	0.86
B76-4	40	60	5	3	1	0.10	2.5	—	0.99
B76-5	40	60	5	3	1	—	2.5	—	2.8
B76-6	40	60	5	3	1	0.05	2.5	—	—
B76-7	40	60	5	3	1	0.05	2.5	—	—
B80-1A/C	40	—	5	1	—	—	2.0	—	—
B80-2/4	40	—	5	1	—	—	2.0	—	16.4
B82-1	40	—	4	1	—	—	2.0	—	—
B82-2	40	—	4	1	—	0.02	2.0	—	—
B82-3	40	—	4	1	—	0.04	2.0	—	—
B82-4	40	—	4	1	—	0.06	2.0	—	—
B82-5	40	—	4	1	—	—	2.0	0.5	—
B82-6	40	—	4	1	—	0.02	2.0	0.5	—
B82-7	40	—	4	1	—	0.04	2.0	0.5	—
B82-8	40	—	4	1	—	0.06	2.0	0.5	—
B84-1	40	—	0	3	—	0.10	2.5	—	—
B84-2	40	—	1	3	—	0.10	2.5	—	—
B84-3	40	—	2	3	—	0.10	2.5	—	—
B84-4AD	40	—	3	3	—	0.10	2.5	—	—
B84-5AD	40	—	4	3	—	0.10	2.5	—	—
B84-6	40	—	5	3	—	0.10	2.5	—	—
B84-7	40	—	6	3	—	0.10	2.5	—	—
B84-8	40	—	8	3	—	0.10	2.5	—	—
B-6	100[a]	—	4	—	4	—	4.0	—	18.5

a 100 parts Ruberoid asbestos floats.

Table 3

MILLING CONDITIONS AND FLOW TIMES FOR DAP FORMULATIONS

Mill Lot No.	Milling Conditions: Milling Time min.	Surface Temp. °C	Roll Spacing in.	Net Input Power KW	Weight After Milling Kg	Cup Flow Times: Before Drying Time sec	Before Drying Pressure tons	After Drying Time sec	After Drying Pressure tons
B73-3A	22.5	88	0.080	2.10	0.85	12.5	5	9.53	10
B73-3B	21.0	89	0.080	2.08	1.10	11.15	5	9.53	10
B73-4	11.0	82	0.040	1.95	0.50	6.78	5	6.72	10
B73-5	14.5	79	0.040	2.02	0.50	6.20	5	7.74	12.5
B73-6	24.5	80	0.080	2.00	0.95	9.90	5	12.50	10
B74-4	55	75	0.176	2.30	1.95	6.64	5	9.76	5
B75-5	34	80	0.160	2.00	1.55	6.66	5	8.00	10
B75-6	45	81	0.160	1.95	1.47	Did not cure		Did not cure	
B76-3	40	77	0.160	1.95	1.40	6.29	5	8.49	5
B76-4	47	—	0.145	2.03	1.40	6.14	5	8.44	5
B76-5	35	77	0.145	2.13	1.50	13.56	5	10.03	10
B76-6	50.5	77	0.145	1.99	1.37	7.13	5	10.01	5
B76-7	54	77	0.186	2.10	1.94	6.03	5	21.48	5
B80-1A	29	76	0.040	2.75	0.45	6.48	5	10.08	5
B80-1B	26	75	0.040	2.32	0.46	4.80	5	9.42	5
B80-1C	35	75	0.080	1.92	0.46	4.83	5	13.76	5
B80-2	95	90	0.160	2.60	1.75	18.00	5	16.83	10
B80-3	67	83	0.176	1.48	1.48	5.39	5	10.16	5
B80-4	63	85	0.125	1.83	1.34	—	—	18.20	5
B82-1	64	74	0.120	1.77	1.00	7.96	5	9.35	5
B82-2	62	67	0.130	1.70	1.35	6.58	5	9.08	5
B82-3	58	68	0.140	1.79	1.40	5.42	5	9.30	5
B82-4	58	74	0.120	1.77	1.15	4.79	5	9.00	5
B82-5	48	70	0.120	1.80	1.10	4.80	5	8.85	5
B82-6	57	67	0.140	1.84	1.57	4.83	5	11.77	5
B82-7	53	74	0.120	1.87	1.14	4.88	5	11.94	5
B82-8	50	66	0.140	1.84	1.45	4.17	5	12.62	5
B84-1	60	80	0.150	1.80	1.26	6.45	5	11.50	5
B84-2	80	80	0.150	1.73	1.40	8.14	5	12.50	5
B84-3	91	75	0.166	1.73	1.40	8.52	5	8.86	5
B84-4	52.5	60	0.100	2.10	1.15		—	8.53	5
B84-5	36	66	0.060	1.80	1.40	—	—	8.39	5
B84-6	76	69	0.080	1.60	1.35	4.57	5	8.38	5
B84-7	65	65	0.156	1.89	1.45	4.87	5	8.32	5
B84-8	97	71	0.146	1.75	1.50	3.55	5	7.67	5
MB-1938	—	—	—	—	—	11.47	5	17.60	5

Evaluation is incomplete, but an increasing amount of evidence is accumulating to indicate that present evaluation techniques are not adequate.

It has been felt that residual volatiles in the DAP molding powder may result in cracks and voids in moldings. It has been shown that increased volatile content results in an increase in porosity of Flow Test Cups. Some additional data on volatile content after milling and after drying are given in Table 4.

Material milled to a longer flow time tends to have a lower volatile content, but even material milled to be much too stiff still has too high a volatile content. Drying is therefore essential. The relatively high volatile content of some of the resins after drying is probably the result of insufficient stirring during the first few hours of drying. The relatively low loss of volatiles upon drying of Mesa B-1938 can probably be explained by the fact that Mesa DAP contains larger particles than does Mound Laboratory DAP.

Table 4

VOLATILE CONTENT OF DAP AFTER MILLING AND AFTER DRYING

Sample Number	Volatile Content, Per Cent	
	After Milling	After Drying
B21	–	0.29
B73-3	1.77	–
B73-4	2.25	0.88
B73-5	2.35	0.78
B76-3	–	0.66
B76-4	–	–
B76-5	–	0.50
B76-6	–	0.46
B76-7	–	0.70
B79-1	–	0.59
B80-1	1.92	–
B80-2	1.67	–
B80-3	1.53	–
B80-4	2.07	–
B81	1.292	–
B83	3.39	–
Mesa B-1938	1.10[a]	0.70

a As received at Mound Laboratory

Evaluation A relatively large number of samples have been submitted to the Production Division for molding and evaluation. Evaluation has been slow, and in some cases, there has been some difficulty in positively determining to which material certain evaluation data pertains.

Data for the first X-ray inspection and for cure are given in Table 5, while data for the second X-ray inspection after thermocycling are given in Table 6.

Table 5

FIRST X-RAY INSPECTION OF MOUND LABORATORY DAP FORMULATIONS

Sample Number	Number X-rayed	Cracks %	Voids %	Inclusions %	Cracks Plus Voids %	$CHCl_3$ cure min
B6	25	0	8	68[a]	8	18.5
B73-3	25	0	0	4	0	11.2
B73-4	25	0	0	4	0	20.1
B73-5	25	0	0	4	0	7.6
B73-6	25	0	0	4	0	b
B74-4	25	0	60	4	60	3.6
B76-4	25	20	4	0	24	0.99
B76-5	30	0	13	0	13	2.8
B76-6	25	4	8	4	12	b
B76-4AD	25	0	44	4	44	3.55
B76-5AD	25	0	20	16	20	14.8
B76-6AD	25	4	28	0	32	3.6
B76-7AD	25	12	44	8	56	2.2
B80-2	25	28	8	0	32	16.4

a B6 was made from high magnetite content asbestos.
b Data not yet available.

Table 6

X-RAY INSPECTION OF MOUND LABORATORY DAP AFTER THERMOCYCLING

Sample Number	Cracks %	Voids %	Inclusions %	Cracks plus Voids, %	
				2nd X-ray	1st X-ray
B6	0	8	68[a]	8	8
B73-3	100	0	0	100	10
B73-4	100	0	0	100	0
B73-5	100	0	0	100	0
B73-6	100	0	0	100	0
B74-4	80	20	0	100	0
B76-4	b	b	b	b	24
B76-5	100	0	0	100	13
B76-6	80	8	0	88	12
B76-4AD	92	8	0	100	44
B76-5AD	100	0	0	100	20
B76-6AD	64	32	0	96	32
B76-7AD	56	40	0	96	56
B80-2	92	8	0	100	32

a B6 was made from high magnetite content asbestos.
b Data not yet available.

Various samples are compared on the basis of percentage of cracks plus voids. All heads, including those with inclusions, were examined for cracks and voids; hence, the figures for cracks and voids are the true totals of such defects.

Although a number of samples had a fairly low rejection rate on the first X-ray, all were very poor on the second X-ray except for B6, which is material which appeared to have poor molding characteristics when first made. It has aged for approximately eighteen months.

The B73 materials were all very good on first X-ray inspection, but all gave 100 per cent cracks after thermocycling. These materials were so stiff that the absence of cracks on first X-ray was somewhat surprising. It is possible that if these materials had been milled to a proper flow time, there would have been much less cracking during thermocycling.

It has been felt for some time that the molding conditions presently used for production molding are not optimum for Mound Laboratory DAP, and, more recently, that they might not be optimum for Mesa DAP either. A series of tensile test dog-bone bars were molded from Mesa B-1938 DAP using several temperatures and preheat times. The data are incomplete since the Wabash press failed before all of the bars were molded. However, the data obtained to date are of interest and are presented in Table 7.

Test bars molded at 170°C were very poor in appearance and gave low to very low tensile strengths. All test bars molded at 140°C had good physical appearance and had good tensile strength. Nine out of ten test bars molded at 160°C had good appearance and had good tensile strength.

It would appear that 160°C is probably near the upper limit for obtaining good moldings, while 140°C may be near the lower limit unless longer curing times are used. More definite conclusions cannot be made until more complete data are available.

Table 7

EFFECT OF MOLDING CONDITIONS ON TENSILE STRENGTH OF MESA B-1938 DAP

Preliminary Data

Mold Temp °C	Preheat Time min	Appearance of Samples	Type of Breaks No. Good	Type of Breaks No. Bad	Thickness at flat, in.	Tensile Strength psi
170	3.0	poor	1	–	0.145	4795
170	2.5	poor	1	–	0.129	6550
170	2.0	poor	2	1 ?	0.136	3370
170	1.0	poor	–	1	0.142	1520
160	2.0	good	3	–	0.129	8210
160	1.0	good	1	–	0.129	7675
160	0.5[a]	good[b]	3	1	0.126	8390[c]
160	0.5[a]	good	2	–	0.117	9190
140	2.5	good	1	–	0.124	7660
140	2.0	good	3	1 ?	0.123	8790
140	1.0	good	1	–	0.115	8090

a Run on dried material; others run on material as received.
b Specimen giving bad break had poor appearance.
c Low value of sample giving bad break omitted.

Some of the bars made in the preliminary work on the Mound Laboratory flow test had filled the mold sufficiently that the resultant partial dog-bones could be held in the tensile-test jaws. The tensile strength data obtained with these bars are given in Table 8.

Table 8

TENSILE STRENGTH OF TEST BARS MADE

IN MOUND LABORATORY FLOW TEST

Molded at 148°C and ~95 psi pressure

Sample	Thickness at Flat, in.		Tensile Strength, psi	
Number	No Preheat	Preheat	No Preheat	Preheat
B21-1	0.177	-	655	-
B79-1	0.200	0.217	2050	4020
B79-1	0.160	-	3250	-
B79-2	-	0.140	-	5715
B79-3	0.228	0.133	1405	4435
B79-4	0.206	0.127	1990	5510
B79-4	-	0.125	-	2560
B79-5	0.196	0.128	2400	4455
B79-6	0.226	0.129	1150	4420

Some surprisingly high values were obtained on bars made from preheated preforms, and in all cases preheating resulted in higher values of tensile strength.

Determinations of physical properties on test specimens molded under our standard conditions are summarized in Table 9.

The test specimens for Mesa B-1938 were molded from material which had been dried in the circulating-air oven. It is probable that the resultant reduction in volatile content somewhat enhanced the tensile strength and had an even more marked effect on the impact strength.

Table 9

PHYSICAL PROPERTIES OF DAP FORMULATIONS

Sample Number	Tensile Strength psi	Izod Impact Strength ft - lb per in. Notch	Heat Distortion Temp. °C	Arc Resistance sec
B71-2	7115	-	142	132.6
B73-3	6550	0.280	152	-
B73-4	5150	0.280	151	132.9
B73-5	7775	-	-	138.9
B73-6	7085	0.291	151	134.6
B74-2/3	7040	-	151	132.9
B75-1/2	6960	-	144	133.3
B75-3	6690	-	-	130.5
B75-5	-	0.277	148	132.0
B76-1	7970	-	159	131.8
B78-1	7835	-	150	130.7
B78-2	6660	-	145	128.5
B79-1	9425	-	153	29.2
B79-2	-	-	-	36.5
B79-3	9045	-	-	35.3
B79-4	7360	-	-	33.5
B80-2	8125	0.245	154	-
Mesa B-1938[a]	8690	0.295	158	134.9
Acme CF[b]	-	-	-	107.7

a Test bars made from oven-dried material.

b A cellulose-filled DAP

Additional work has been done on the Mound Laboratory flow test. Test temperatures of 140°C and 160°C were used, and an air pressure of 60 pounds per square inch was adopted. Approximately 50 pounds per square inch is required to insure positive movement of the press platen, so 60 pounds per square inch is about the lowest pressure which can be expected to give consistent results.

The numerical data obtained for 16 DAP formulations are given in Table 10. The cup flow test results are included for comparison.

Table 10

MOUND LABORATORY FLOW TEST-AIR PRESSURE ON WABASH PRESS 55 TO 60 PSI

Sample Number	Time to Close at 140°C sec	Time to Close at 160°C sec	Amount not Closed at 140°C in.	Amount not Closed at 160°C in.	Creep at 140°C in.	Creep at 160°C in.	Flow time at 5 tons sec
Mesa B-1860	174	160	0.317	0.398	0.0037	0.001	11.51
PB1	152	118	0.347	0.375	0.0061	0.004	10.30
PB2	145	105	0.320	0.484	0.0063	0.005	11.42
B79-3	128	85.5	0.168	0.310	0.0017	0.006	9.03
B79-4	134	99	0.107	0.294	0.002	0.005	10.37
B80-2	246	192	0.483	0.519	0.009	0.009	16.8 (at 10T)
Mesa-1938	157	115	0.334	0.316	0.003	0.0015	11.47
B73-3A/B	178	143	0.462	0.421	0.009	0.005	9.5 (at 10T)
B84-1	120.5	81.5	0.144	0.234	0.004	0.007	11.50
B84-2	122.5	88.5	0.144	0.207	0.002	0.003	12.50
B84-3	120.0	70	0.092	0.200	0.004	0.006	8.86
B84-4	119.0	73	0.073	0.210	0.0015	0.006	8.53
B84-5	115.0	69	0.053	0.189	0.002	0.006	8.39
B84-6	106	58	0.0355	0.210	0.0015	0.006	8.38
B84-7	119	60	0.038	0.198	0.003	0.006	8.32
B84-8	95	56	0.012	0.164	0.003	0.006	7.67

It is apparent that there is no correlation between the two types of flow test, and that the Mound Laboratory flow test reveals marked differences among materials which give essentially the same flow times when measured by the cup flow test.

It has not yet been determined if numerical data obtained in the Mound Laboratory flow test can prove useful in predicting molding characteristics, but visual observation of the test specimens produced by this test has been informative.

In general, test specimens made at 160°C show evidence of less smooth flow and are obviously more porous than test specimens made at 140°C. In most cases, the ends at which the preform was originally placed show depressions after cooling when a temperature of 160°C is used, but show no, or materially reduced, depressions when a temperature of 140°C is used. Materials which give good flow and which are probably composed of porous grains, give test specimens which have a sponge-like structure when made at 160°C but have a markedly more normal structure when made at 140°C.

Some work is being done in an attempt to evaluate the relative rate of hardening of DAP formulations under heat, and the effect of temperature on the rate of cure. The following technique is being used. A 1⅜ inch diameter preform is made from two grams of material. This gives a wafer 0.065 to 0.070-inch thick. The wafer is placed on a heated metal surface which is held at constant temperature, and the time required for the top surface to become impenetrable to a probe having a sharp point is measured. Preliminary measurements made at relatively high temperatures are given in Table 11.

Table 11

SET-UP TIME OF DAP vs TEMPERATURE[a]

Sample Number	Seconds at 304-308°C	Seconds at 266-269°C	Seconds at 217-219°C
B79-1	36	39	59
B79-2	37	32	51
B79-3	35	33	56
B79-4	38	40	63
B79-5	35	48	65
B79-6	39	39	57
Mesa B-1938	28	—	40

a Time for top surface of a 2-gram preform, 1⅜ inches in diameter and 0.065-0.070 inches thick, to become impenetrable to a sharp probe.

There appears to be little real difference among the various members of the B-79 series, but there may be a significant difference between the B-79 series and Mesa B-1938. Additional work will be required before the possible usefulness of such a test can be determined.

It has been felt for some time that the chloroform extraction test did not give an accurate picture of the true amount of cross-linking and hence of cure. It would appear that an increase in porosity will give a shorter cure time value even though the degree of cross-linking is essentially the same.

Thus, heads molded from B73-4 and B73-5 before they were dried gave cure values of 0.99 and 2.8 minutes, but heads molded from these same materials after they were dried gave cures of 3.55 and 14.8 minutes. Since an increase in porosity might well increase the surface area in contact with chloroform, and since the extraction is carried out for a limited period of time, the data obtained with heads molded from the B73 series are not unexpected.

A variation in the content of non-DAP materials soluble in chloroform could also result in variation in the chloroform cure value. Semiquantitative determinations of the solubility in chloroform, at room temperature, of some of these materials has been made. Since Acrowax appeared to be difficult to disperse readily, its room temperature solubility in some other solvents was also determined. These solubilities are summarized in Table 12.

Table 12

SOLUBILITIES OF POSSIBLE DAP COMPONENTS AT ROOM TEMPERATURE

Material	Grams in 100 Grams of Solution			
	Varnish[a]	Acetone	$CHCl_3$	Ethanol
Acrowax	0.25	3.26	3.58	4.40
Zinc Stearate	–	–	0.187	–
Calcium Stearate	–	–	0.094	–

a A 50% solution of Dapon-35 in acetone

Acrowax appears either to be a mixture of two or more compounds which have different solubilities, to contain a rather wide range of molecular weights, or both.

It would appear that change in internal lubricant or change in concentration could affect the chloroform cure value without actually affecting the degree of cross-linking.

Bridge-Wire Metallurgy *An investigation of interalloy effects between gold and solder has been begun in order to obtain information which may be of benefit in minimizing wire failures in 1E26 detonator heads.*

Physical Testing A concentrated effort has been directed at developing an explanation for the random deformation of bridge wire assemblies observed at the various temperatures. Previous experimental results indicate that the effectiveness of tinning the gold wire (prolonged soldering time) may have an effect upon the deformation. The possible causes considered include segregation of the elements within the solder, variation of the amount of heat applied in soldering the bridge wire, and imperfections in the surface of the gold wire caused by either mechanical or chemical means.

Segregation within the solder was investigated with twelve bridge wire assemblies constructed using indium metal alone as the solder. The twelve heads were subjected to the treatment which would normally produce the previously described deformation phenomenon. There were no deformations, but there was an appreciable reaction between the gold wire and the indium. Serious embrittlement of the gold wire at the interface of the gold wire and mound was observed in each case.

A controlled heat study was made with twelve assemblies constructed with the normal lead-tin-indium solder. They were subjected to a temperature of 185°C for twelve minutes to establish controlled soldering conditions. The thermal treatment that normally produces deformation did not increase the incidence of deformation appreciably, in this case.

The third factor investigated was the presence of imperfections in the surface of the gold wire. A length of gold wire was abraded slightly with 6/0 carborundum paper. Another length of gold wire was etched by a ten-minute immersion in aqua regia. These two sections of gold wire were used in the construction of bridge wire assemblies which were subsequently thermally treated at conditions which normally cause the deformation phenomenon. The results showed that 80 per cent of the wires underwent deformation regardless of the method of surface treatment.

The above results suggest that imperfections on the surface of the gold wire at the interface can consistently cause a deformation of the bridge wire. The random deformation phenomenon which had been previously observed may well be attributed to very minor imperfections, caused during manufacture or handling, which would, on occasion, be located at the interface of the solder mound and gold wire. Effective tinning of the gold wire appears to enhance the probability of the occurrence of deformation of the bridge wires which contain surface imperfections.

Further investigation of the relationship of imperfections in the gold wire to deformation of the bridge wire assemblies is in progress. To eliminate the formation of $AuIn_2$, which has been observed to be present in all deformation cases, bridge wire assemblies using abraded gold wire and 60 per cent tin and 40 per cent lead solder are being constructed. The use of the scratched wire is expected to accent any possible reaction between the 60-40 solder and gold wire.

Bridge wire assemblies, which have been under moisture and heat treatment for several months, are being test fired to determine the effect of severe bridge wire deformation and conductivity changes upon the firing performance. Untreated, normal bridge wire assemblies without explosives have been fired to provide a standard for evaluation. Two boards of 28 untreated assemblies each were fired in the same manner as is used for production heads, using 1E15 bridge wire assemblies as references. It was found that the average T_m for one board was a negative 0.06 microseconds with a variation of 0.110 microseconds to 0.010 micro-

seconds. The second board gave an average negative T_m of 0.08 microseconds with a variation of 0.114 microseconds to 0.043 microseconds. Not one positive T_m was obtained.

No correlation of firing time with the conductance values of these normal assemblies could be established. The close agreement of the average T_m and median T_m attests to the reliability of firing of these groups of 1E26 bridge wire assemblies. It is recommended that T_m and sigma of 1E26 production units containing explosives be compared to 1E26 bridge wire assemblies instead of 1E15 assemblies in order to prevent the intorduction of the observed negative factor into the final T_m values.

Metallographic and X-ray Studies A series of specimens were prepared and polished so that the interface between the bronze electrode and the lead-tin-indium solder could be examined by metallographic techniques. Each specimen showed a surface reaction only. There was good bonding between the electrode and the solder. However, specimens which had been stored for two months at 140°C before mounting showed an extensive reaction between the solder and the cladding material on the electrode. This resulted in a new metal phase which extended along the electrode wire to a depth of 0.012 inch. However, bridge wire assemblies which had not been stored under such severe thermal conditions that complete distruction of the bridge wire had not occurred, showed no such interaction between the solder and the cladding material. Therefore, it is concluded that the present indium-bearing solder will not have detrimental effects on the electrodes.

Examination of the bonds between the bronze electrode wires and the bismuth-gold-lead solders indicated that acceptable wetting of the bronze does not occur with this solder. Therefore, investigation of this solder has been discontinued.

Solders containing bismuth, gold, and tin are being investigated as possible substitutes. The composition of the ternary eutectic mixture is 40 weight per cent tin, 58 weight per cent bismuth, and 2 weight per cent gold, and has a melting point of 134°C. Several different mixtures of these three metals have been prepared up to a maximum gold content of 7 weight per cent. These ternary mixtures are estimated to be in the valley connecting the ternary eutectic to the gold-tin eutectic which contains 10 weight per cent gold. Three test lots of 1E26 bridge wire assemblies using three of these solders have been prepared and are being tested. The first has the ternary eutectic composition, the second has 7 weight per cent gold, and the third has 5 weight per cent gold. These solders wet the bronze electrodes satisfactorily. The solderer reported the handling properties of these solders to be good. Conductivity measurements indicated the resistance to be in the same range (0.031 to 0.034 ohms) as that of the production solder presently used.

Specimens were examined by metallographic techniques to determine any reactions that may take place between the gold bridge wire and the bismuth-tin-gold solder during soldering operations. One solder used was the ternary eutectic mixture and the second solder had the composition of 27.7 weight per cent bismuth, 64.7 weight per cent tin, and 7.7 weight per cent gold and a liquidus temperature of approximately 200°C. The second solder was selected on the basis of its liquidus temperature which is considered to be the top temperature at which normal soldering techniques can be used. Half of the mounds with the gold wire in place were reworked several times by the solderer. All specimens were mounted, cross sectioned through the gold wire, polished, and examined at 500X magnification.

Evidence of a slight reaction of the gold with the solder was observed in the lots using the ternary eutectic solder. No observable difference was observed between the normally-soldered assemblies and the assemblies which had been reworked during the soldering operation. The reaction produced a homogeneous material appearing as a single phase. This phase appears to be the result of a slight dissolution of the gold in the solder matrix without a sharp boundary.

Examination of the systems made from the higher-melting bismuth-gold-tin solder mixture revealed an appreciable loss of gold wire by solution in the solder. The polished metallographic samples showed a loss of 35 to 40 per cent of the gold wire for the normally soldered assemblies and a complete solution of the gold wire in the solder in the reworked assemblies. The material formed as the wire partially or completely disappeared appeared to be the same as that formed in the ternary mixture except that in the latter cases it extended throughout an extensive portion of the solder mound. No grain boundaries were observed. The embrittlement obtained when the indium-lead-tin solder is used and $AuIn_2$ is formed was not found in this case. It is believed that the major cause of the extensive loss of the gold wire in the solder is the higher heat required for the higher melting solder.

Helium-3 Recovery Project *Mound Laboratory has been given the responsibility of recovering helium-3 from Savannah River Plant gaseous wastes. A process involving both chemical and isotopic purification is being developed.*

The palladium diffuser was put in place in the gas purification hood. Water-cooling lines were connected, and a pyrometer to read the temperature of the palladium coil was installed. An ammeter is being installed in the electrical supply line. When this is done, the electrical system will be complete. Heavy-wall Tygon tubing will be used for gas transfer in test runs until enough information is at hand to complete the copper tubing manifold.

Tritium Recovery Project *A process is to be developed for the unloading of tritium reservoirs, the chemical and isotopic purification of the tritium, and the reloading of the reservoirs. The process is to include the capability of loading and unloading at special high pressures. The initial process development will include the recovery of tritium from Los Alamos Scientific Laboratory's decayed salt, and from aqueous and gaseous materials.*

The Los Alamos Scientific Laboratory tritium materials were received and are being stored in an area which had been previously modified for this purpose. The lithium deuteride and tritide salts will be held in storage until the Hot Gas Facility is ready for operation. The gaseous contents of some of the twelve-liter stripping containers will be processed in existing facilities because of their low tritium content.

One of the deuterium gas transport trailers was received. This trailer will be used as a hydrogen storage tank in conjunction with the operation of the low temperature distillation column.

The lead decomposition or amalgamation process will be used for the recovery of tritium from the Los Alamos Scientific Laboratory storage cans and old salt reactors containing unreacted Li(D,T). This is the process used successfully at Los Alamos for the determination of free lithium, deuterium, and tritium in Li(D,T). The method was developed and used previously be Bergstresser and Waterbury of CMR-1 (LAMS-1698).

The decomposition process as it is to be used at this site consists of the following steps:

1. Sample Preparation.

The dry Li(D,T) salts are to be handled only in an argon atmosphere contained in an air-tight, steel drybox. The samples will weigh approximately 75 grams each and will be accurately weighed on a Mettler semi-quantitative balance located inside the drybox. A maximum weight of 75 grams per sample has been selected because of the limited capacity of the standard gas containers (90 liters).

2. Outgassing.

Prior to the decomposition of the tared Li(D,T) sample with lead, it is necessary for the sample to be degassed on the process line under vacuum. The loaded reactor vessel is heated to approximately 750°C, and the gases, principally helium-3, are collected in standard volume containers. The outgassing procedure effectively eliminates possible gas-pockets in the salt that could result in an explosion similar to the one that occurred at Los Alamos Scientific Laboratory in February, 1956.

3. Lead Decomposition.

When the reactor vessel has cooled following the outgassing operation, it is returned to the drybox, opened, and a prescribed amount of lead metal is added. The correct Pb/Li(D,T) ratio, as reported by Los Alamos Scientific Laboratory scientists, is 12 to 1. If the ratio is less than 12 to 1, the chemical reaction is incomplete and results in the presence of unreacted Li(D,T); an excess causes the melt to become very brittle. The lead is added to the reactor in the form of "pigs". These are purified metal casts that have been formed by melting known quantities of reagent-grade lead metal in small nickel or graphite crucibles. The dross is skimmed-off prior to hardening.

With the addition of lead, the reactor vessel is closed, returned to the evacuated process line, and placed in operation. Controlled heat is applied and the temperature in the reactor slowly raised to 300-400°C. Most of the gases are liberated from the Li(D,T) at this temperature. When the evolution of gas has nearly subsided, additional heat is applied until a temperature of 750°C is reached. The elevated temperature succeeds in driving off the final traces of $(D,T)_2$. This step also helps to clean the upper walls of the of the container of froth and condensate which may have collected during the vigorous evolution of gases.

The Collins Helium Cryostat was received at Mound Laboratory during January, 1958 and is being stored pending modification of the SW-1B Area. A 4-6 inch layer of concrete is to be poured over the present SW-1B floor. The piping and manifold used to bring the make-up hydrogen into the building is being installed. This will permit the hydrogen cylinders also to be located in the security area just south of the building.

Specifications for the helium manifold have been given to the Engineering Division. Specifications for the hydrogen system have been written and the flow sheet is being drawn.

Eight runs have been made on the low temperature still with the system oxygen-16/oxygen-17/oxygen-18. Various vapor velocities were used. Analysis of the samples indicates no significant separation of the oxygen isotopes to date.

The installation of calorimetry in the Hot Gas Facility assay room is now complete except for control units. This will be a single unit and is being designed and made up by the Electronics Department. It will include an amplifier and chopper circuit in one compact instrument. Several alterations have been made to the bath itself in an effort to improve its operation.

The bath has been continuously stirred for several days for the purpose of temperature equilibrium studies.

Isotope Separation Project *Mound Laboratory is responsible for the production of several isotopes needed in research projects at several Division of Military Application sites. Particular emphasis is placed on the investigation and use of the thermal diffusion process for separating gaseous isotopes. Processes and facilities to handle both inert and radioactive gases are being developed.*

Gaseous Thermal Diffusion The continuous-flow separation of neon isotopes in the four-column series cascade has been completed. A total of 5.2 liters (STP) of enriched neon-22, having an isotopic purity of approximately 90 per cent, was collected first. This gas has an air contamination of less than 0.2 per cent. The neon-20 residues of this separation were reprocessed, and approximately 42 liters of neon-20 with an isotopic purity of better than 99.8 per cent was collected. At present, this gas is contaminated with about 30 per cent air.

Following this separation, the connecting lines between hoods were removed, and further evaluation of certain thermal diffusion factors are being investigated on the 12-foot long, 1.61-inch diameter column with the hydrogen-deuterium system. The thermal diffusion gas transport coefficient, H, for hydrogen-deuterium mixtures is being more precisely determined. This work will be continued until the hood and columns are needed for the proposed helium isotope separation program.

Very pure samples of hydrogen and deuterium have been prepared using the two thermal diffusion columns in the south hood. These samples are being used to determine fragmentation ratios of these gases in the Model 21-201 mass spectrometer. The south hood is to be used for preliminary separations of noble gases when this program is activated.

Concurrent with above work, the gas handling systems in both hoods are being thoroughly leak-tested.

Design work on the proposed hot gas facility has been completed, and the construction contracts have been placed out for bid. In addition, purchase orders for equipment have been initiated, and some of the equipment has started to arrive.

The Consolidated Model 21-620 mass spectrometer was placed in operation in room R-108 by a field engineer from Consolidated Electrodynamics Corporation who indicated that the ionizing potential on the ion source is too low-only about 23 volts as opposed to a recommended value of 70 volts. This difference is due to factory installation of a silicon diode of improper rating. As the ionizing potential affects both the sensitivity of the instrument and the fragmentation patterns of various molecules, the final calibration of the instrument is awaiting arrival of a replacement silicon diode.

In order to analyze xenon isotopes more precisely, two additional changes have been incorporated in the instrument. The automatic sweep-scanning rate was slowed somewhat by the addition of a 100 megohm resistor to the capacitor bleed control in the mass selector potential circuit. With this modification the xenon isotope mass range is now scanned in about 36 seconds instead of 24 seconds. In addition, the Brown recorder originally installed by Monsanto in the instrument has been replaced with an L. and N. Speedomax, which has a significantly faster response time. The replacement of the recorder also has reduced the background noise in the instrument to some extent.

A total of 33 neon samples, 42 hydrogen-deuterium samples, and 40 oxygen samples have been analyzed on the Model 21-201 spectrometer this month. This work was done in conjunction with the above experimentation and in conjunction with experimentation on the low temperature distillation column.

Liquid Thermal Diffusion To continue the study of the effect of time on separation factor for the H_2O-D_2O system, a 168-hour run was made, again employing a 50-50 volume per cent mixture as feed and a temperature difference between walls of 17°C. The separation factor, based on the end fractions, followed the same time trend as that exhibited by previous runs with the same feed and temperature difference. This trend may be summarized in the four separation factors obtained with different times, namely: 48 hours, 1.034; 96 hours, 1.050; 168 hours, 1.085, and 239 hours, 1.156. A plot of these data gives a smooth curve with continually increasing slope, suggesting that equilibrium has not yet been approached. Separation factors for the different times, based on fractions removed from the ends, exhibited several discrepancies which have not yet been resolved.

The study of the effect of D_2O concentration in the feed on separation was extended with a run employing a mixture containing 75 per cent by volume of D_2O as feed. The temperature difference was again 17°C and the time 48 hours. The separation factors, based on the end fractions, from this new run and two previous runs made with the same time and temperature difference but with varying concentrations of D_2O in the feed, indicate that separation increases with increasing concentration of D_2O in the feed. Thus with 25, 50, and 75 per cent D_2O in the feed, factors of 1.000, 1.034, and 1.052, respectively, have been obtained. However, no consistent separation was observed in the other fractions under these conditions.

A run, employing as feed a TBP solution approximately 0.05-molar in cerium nitrate and 0.05-molar in yttrium nitrate, a temperature difference of 41°C, and a time of 48 hours, has been completed. As in previous runs with more concentrated feeds, a gross concentration of solutes in the bottom fractions occurred. However, the mole ratio of cerium to yttrium for the different fractions did not exhibit any consistent difference from the value for the feed. The liquid phase separation, which had been observed in the bottom fractions in runs employing more concentrated feeds, was reduced in amount but was not eliminated.

In an effort to eliminate phase separation completely, two volumes of the TBP feed, nomially 0.1-molar in each salt, was diluted with one volume of ethanol, thus reducing each salt concentration to approximately 0.065 molar. Runs of 48, 96, and 234 hours duration have been completed with this new feed. The average temperature difference was again 41°C. The dilution of the feed with alcohol eliminated phase separation completely. Complete analytical results are available only for the 48 and 96-hour runs. These show no gross separation of solutes from solvent in the 48 hour run and only a weak tendency to concentration of the solutes toward the bottom of the column in the 96-hour run. No separation of cerium from yttrium was observed in either run.

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.*

In last month's report, successful results were reported for the recovery of plutonium added to 24 urine samples from 20 different donors. The counting level was 243 alpha counts per hour as compared with a maximum permissible concentration (MPC) of 210 counts per hour. The average plutonium recovery was 93 per cent.

To test the sensitivity of the method, a series has been run in which the counting rate of the added plutonium was 27.5 alpha counts per hour per 24-hour sample. Eight samples were obtained from six donors. Four of the samples were collected over a 48-hour period, and half of each of these was used as a control to determine the personal background of the donor at the time the sample was taken. The other four samples were corrected for personal background from the routine urinalysis records. The average recovery of plutonium was 91 per cent (Table 1).

The procedure is now believed to be satisfactory for routine use. The modification of the cerium phosphate precipitation step (described in last month's report) is being adopted in the routine determination of actinium and thorium as well. It is hoped that, in the near future, additional data will be obtained to determine if a minimum amount of calcium phosphate precipitate is necessary for quantitative yields.

Table 1

RECOVERY OF PLUTONIUM FROM HUMAN URINE

Donor	Urine Volume ml	Ca Ppt. Volume ml	Yield cts/hr	Background* cts/hr	Net Recovery cts/hr	Net Recovery %
1	1980	150	31.88	7.15[a]	24.73	89.9
2	1260	100	32.86	8.07[a]	23.79	86.5
21	1300	80	32.93	7.00[a]	25.93	94.3
22	550	120	29.63	4.80[a]	24.83	90.3
2	1300	140	32.65	5.59[b]	27.06	98.4
1	1200	200	32.55	5.65[b]	26.90	97.8
4	1150	240	29.53	5.76[b]	23.77	86.4
23	1050	260	35.18	11.81[b]	23.37	85.0

* Personal background corrected for instrument and mounting disk backgrounds.
a Personal background from routine urinalysis records.
b Personal background from a simultaneous determination on one half of a 48-hour urine sample.

Materials of Weapons Interest *Process development for the production of small amounts of unusual materials is occasionally required to meet the needs of the weapons program.*

Ionium Project *The extraction of ionium from partially-processed raffinates from the Mallinckrodt Uranium Refinery is being completed to fulfill previous commitments.*

The concentrated, impure ionium product solution remaining after the shipment which was made to University of California Radiation Laboratory last month, contained approximately 21.5 grams of ionium by alpha count. Two 25-milliliter aliquots of the product solution were adjusted to 3-normal and 4-normal in aluminum nitrate, respectively, to determine whether or not emulsions would be formed upon contact with organic solvents, and to determine extraction efficiencies. In order to lessen the possibilities of introduction of impurities which might cause emulsions, xylene was used as a diluent for the TBP instead of the kerosene-base Amsco solvent used in previous work. The organic solvents were mixed on a 50 per cent by volume basis and were then washed three times each with sodium carbonate solution, water, and dilute nitric acid.

The 4-normal aluminum nitrate solution gave slightly better extraction efficiency. However, the 50 per cent increase in solution volume required to reach this normality involved more time and effort to obtain the same total extraction that could be accomplished with an additional extraction stage of the 3-normal aluminum nitrate solution. Since no emulsion problems were encountered in these test runs, it was decided to make the remaining concentrated product solution 3-normal in aluminum nitrate.

In an aqueous solution containing aluminum nitrate as a non-extractable salting agent, a decrease in acidity increases the distribution coefficient (O/A). Since nitric acid is extracted into TBP, use was made of this phenomenon. As in the test-extractions, no attempt was made to adjust the acidity of the aqueous solution, which was 5.9-normal in nitric acid before addition of aluminum nitrate.

Data on the extraction runs are tabulated below.

Table 1

EXTRACTION OF IONIUM FROM 3-NORMAL ALUMINUM NITRATE SOLUTION

Flow ratio O/A = 0.2

Extraction Number	Ionium in Aqueous g/liter	Ionium in Organic g/liter	Distribution Coefficient
1	5.05	6.46	1.28
2	3.35	7.04	2.10
3	1.93	6.21	3.22
4	0.75	4.43	5.92
5	0.14	2.61	18.60
6	0.034	1.47	43.40

Since the final aqueous phase contained only 0.11 gram of ionium, it was decided to discontinue extraction to prevent loading the organic phase with rare earths and other extractable impurities.

The entire organic phase was scrubbed with a solution prepared from equal parts of 6-normal nitric acid and 2-molar aluminum nitrate. The scrub was used to remove rare earths and other contaminants which may have been extracted into the organic phase. The flow ratio was 10 (O/A). The aqueous scrub solution subsequently found to contain 0.289 gram of ionium, was, in turn, scrubbed with 50 per cent TBP in xylene. The organic-to-aqueous flow ratio was 3. The depleted aqueous scrub solution contained 0.016 gram of ionium and was discarded. The organic scrub solution was added to the main body of organic extractant.

The organic phase was stripped with 0.1-normal nitric acid, and with distilled water. The final stripped organic contained 0.073 gram total ionium and a considerable amount of the initial uranium of the concentrated product solution. The aqueous phase, containing most of the ionium and some of the uranium, was precipitated with ammonium hydroxide. The precipitate will be filtered and dissolved with nitric acid.

The 1.66 grams of scavenged ionium to be purified separatly will be added to the product nitric acid solution, and the uranium will be removed. Finally, this uranium-free ionium solution will be combined with approximately 2.5 grams of ionium to be stripped from TTA solutions to give a final product for rare earth and common element analyses.

Uranyl nitrate solutions collected from pilot plant operations have been sent to Mallinckrodt Chemical Works.

Protactinium Project *The production for Albuquerque Operations of a gram of protactinium-231 from unprocessed Sperry-Filter-Press residues from Mallinckrodt is being completed.*

A spectrographic analysis of the composite FY 1956 protactinium solution has been made (Table 1). The sample consists of all the material precipitable with ammonium hydroxide from the di-isobutyl carbinol solution.

Table 1

SPECTROGRAPHIC ANALYSIS OF COMPOSITE PROTACTINIUM SOLUTION

Element	Per Cent*
Si	0.347
Au	0.047
Mg	0.071
Fe	1.69
Ni	0.087
Pt	0.081
Al	0.159
Nb	3.73
Cu	0.048
Ag	0.025
As	3.86 (?)

* 50%

As expected, the spectrographic analysis showed that niobium and iron are the two principal impurities in the protactinium solution. The presence of arsenic is highly questionable and is being disregarded temporarily. Development of a procedure for separating niobium from protactinium was undertaken first.

Previous work has shown that niobium can be stripped from a di-isobutyl carbinol solution into an aqueous phase which is six normal in hydrochloric acid and five per cent in oxalic acid. Under these conditions, nearly all of the protactinium remains in the organic phase.

Unfortunately, attempts to use this procedure for the present protactinium sample were unsuccessful, possibly because of the high concentration of iron. A variety of mixtures of hydrochloric and oxalic acids were tested, but in all cases, the protactinium was stripped along with the niobium. Although some fractionation occurred, the separation was considered unsatisfactory.

As a second approach, attention was directed towards the separation of iron from protactinium and niobium. If the di-isobutyl carbinol solution is stripped with a phosphoric acid solution, the iron remains in the organic phase, and the protactinium and niobium precipitate out in the aqueous phase. This procedure is satisfactory for batch operations in centrifuge tubes, but, because of the formation of a flocculent precipitate, does not lend itself well to separatory funnel operation. However, in the course of developing a more satisfactory procedure, it is likely that most of the di-isobutyl carbinol will be purified in small batches. Approximately 100 milligrams of protactinium has already been decontaminated of iron and has been concentrated to between two and three milligrams per milliliter in di-isobutyl carbinol.

Development of a method for separating protactinium from its decay products would require more time than is presently available. However, a crude separation of actinium and its decay products from protactinium indicated that less than one per cent of the total alpha activity could be attributed to decay products. This corresponds to an average age of 50 days for the protactinium. Therefore, the concentration of protactinium as reported last month (1.43 milligrams Pa-231 per milliliter of solution) may be provisionally accepted.

Analysis of the protactinium solution on the alpha pulse height analyzer confirms the finding from gamma pluse height analysis that no significant alpha activity is present other than that from protactinium and its decay products.

REACTOR DEVELOPMENT PROGRAM

CIVILIAN POWER REACTORS

Homogeneous Reactors *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 solutions up to 300°C will be measured.*

Table 1 gives a summary of the plutonium (III) and plutonium (IV) phosphate solubilities and the conditions of measurement. The temperature values may be lower than those listed by as much as 10°C. This uncertainty is due to the slowness of formation and the fineness of the precipitate. Room temperature (24°C) solubility measurements were taken after several days standing to allow solutions to come to equilibrium.

Table 1

SUMMARY OF Pu (III) AND Pu (IV) PHOSPHATE SOLUBILITY MEASUREMENTS

System	Solubility at 24°C grams Pu/liter	Results on Heating: Pu Conc g/liter	Results on Heating: Remarks
Pu (III), $PO_4^{\equiv}$, H_2O	0.0132	0.109	Not completely soluble up to 281°C.
Pu (III), $PO_4^{\equiv}$, H_2O,	0.215 *	0.215	Crystals appear at 180°C.
saturated at room temp. with SO_2		0.530	Not completely soluble up to 286°C.
Pu (III), $PO_4^{\equiv}$, 0.847 N H_3PO_4	0.008	0.149	Not completely soluble up to 277°C.
Pu (III), $PO_4^{\equiv}$, 8.469 N H_3PO_4	0.0863	0.0863	Crystals appear at 150°C.
		0.127	Not completely soluble up to 281°C.
Pu (IV), $PO_4^{\equiv}$, H_2O	(Negligible background on counting)	0.195	Not completely soluble up to 250°C.
Pu (IV), $PO_4^{\equiv}$, 0.0847 N H_3PO_4		0.195	Not completely soluble up to 270°C.
Pu (IV), $PO_4^{\equiv}$, 0.847 N H_3PO_4	(Negligible background on counting)	0.390	Not completely soluble up to 297°C.
Pu (IV), $PO_4^{\equiv}$, 8.469 N H_3PO_4	0.174	0.195	Completely soluble at 58°C and started to come out at 134°C. On continued heating crystals of different type came out at 211°C.

* being repeated

Investigations were made on the behavior of plutonium (VI) sulfate solutions containing small amounts of lithium sulfate. This impurity was present because of the necessity for preparing the solutions from lithium plutonium (VI) carbonate. Dilute sulfuric acid was added to an excess of lithium plutonium carbonate, and the supernate was heated to drive off the excess carbon dioxide. The solution remained acid (pH 1.4). The results of heating are shown in Table 2. The precipitates formed between temperatures of 177°C and 230°C and were light yellow. They did not redissolve on cooling and showed other characteristics of

plutonium (IV) basic sulfate. Further evidence of reduction was shown in experiment No. 5 in which the solution acquired the blue color of plutonium (III) on heating to 340°C.

An oxidizing agent, ammonium peroxydisulfate with a trace of silver nitrate as catalyst, was added to a nearly neutral plutonium (VI) sulfate solution in an attempt to stabilize the solution. No precipitate was observed up to the maximum temperature of 340°C, although the solution became brown. The possibility of using an overpressure of oxygen as an oxidizing agent was explored in experiments No. 7, 8, and 9. The slight oxygen overpressure, amounting to about one atmosphere at 24°C, did not significantly change the precipitation temperature, although it seemed to prevent the formation of the blue color.

Modifications are being made on a Beckman Model DU spectrophotometer to obtain a more quantitative examination of the oxidation states of the plutonium, and the effects of a greater oxygen overpressure will be examined.

Table 2

HIGH TEMPERATURE STABILITY OF PLUTONIUM (VI) SULFATE SOLUTIONS

Expt. No.	Pu g/liter	H_2SO_4 Normality	Oxidizing Agent	Phase change °C	Phase change type
1	48.8	~ 0.04	–	230	white ppt.
2	20.7	~ 0.04	–	177	white ppt.
3	4.1	~ 0.04	–	216	yellow ppt.
4	4.1	0.8	–	219	yellow ppt.
5	4.1	4.8	–	>340	blue soln.
6	4.1	~ 0.04	$(NH_4)_2S_2O_8$	>340	sl. brown soln.
7	4.1	~ 0.04	O_2	185	yellow ppt.
8	4.1	0.8	O_2	250	yellow ppt.
9	4.1	4.8	O_2	>340	sl. brown soln.

Plutonium Fast Breeder Reactor *Fast breeder reactors are of interest to the Civilian Power Reactor Program. Plutonium, which has a high neutron efficiency, is being considered for use in reactors of this type. Mound Laboratory has been given the responsibility for acquiring data on some of the proposed fuel systems, and for maintaining technical cognizance of fuel cycle problems associated with the Fast Breeder Reactor Program.*

Plutonium Alloy Research *A project has been initiated for the determination of the density, viscosity, heat capacity, thermal conductivity, and phase equilibria of plutonium metal and plutonium alloys proposed as fuels for fast breeder reactors.*

The design work for all services except ventilation for the plutonium laboratory in room R-159 has been essentially completed. The design work for the ventilation system will be started as soon as possible.

The alpha-hoods to be fabricated for this laboratory have been ordered. After receipt of bids, a member of the Engineering Division visited S. Blickman Inc., and determined that gloved alpha-hoods manufactured by them to Mound Laboratory specifications are satisfactory for a plutonium-handling facility. It is expected that delivery on these hoods will be completed before May 1, 1958. These hoods will be constructed of stainless steel and have "Homolite" windows.

Design and experimental fabrication work is being continued on the apparatus to measure the physical properties and study the phase equilibria of plutonium and its alloys.

DEPARTMENT OF DEFENSE

U. S. ARMY SIGNAL ENGINEERING LABORATORIES (Components Branch)

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

NUCLEAR BATTERY-THERMOCOUPLE 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 optimum shape and configuration for maximum efficiency output are being considered, and prototype thermoelectric generators are being constructed.

Construction has been continued on thermoelectric Generator No. 3 described in last month's report. An exploded view of this generator is shown below.

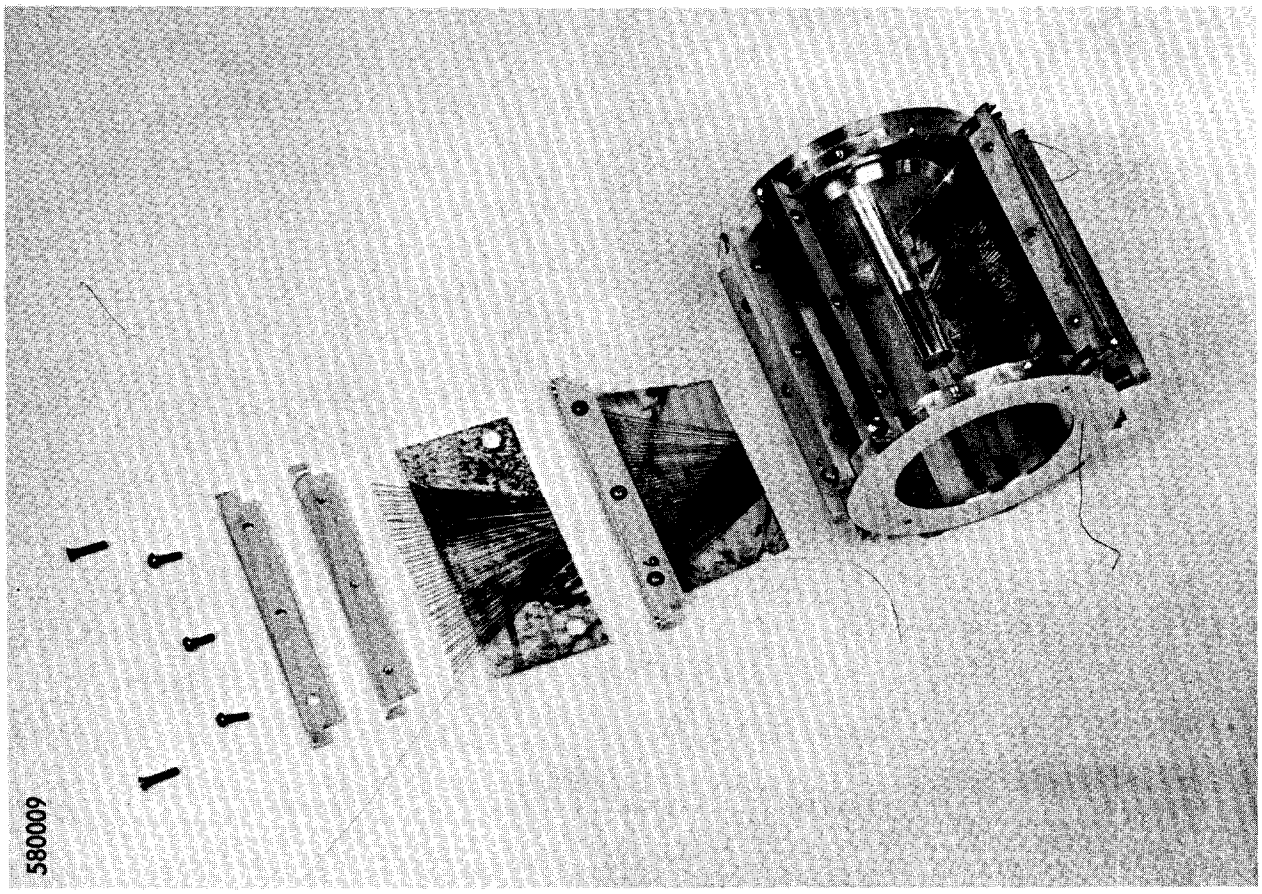

Figure 1. Thermoelectric Generator No. 3