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PLASTICS RESEARCH

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

PLASTICS Sixty mill lots of Dacron-filled diallyl phthalate were made in the Plastic Production Facility. These were combined, at four lots per batch, into 15 batches.

One hundred fifty tensile strength test specimens were run as an evaluation of the 15 batches produced. An average value of 4377 psi was obtained which is acceptable since the tentative tensile strength specification had been set at 3000 psi. The results are given in Table 1. Of the specimens tested, 23 gave "bad breaks" indicating a flaw in the test bar.

Table 1

TENSILE STRENGTH OF DACRON-FILLED DAP

Sample No.	Tensile Strength psi
79-82	4459
83-86	4477
87-90	4371
77-78	4241
91-92	
93-96	4355
97-100	4366
101-104	4692
105-108	4230
109-112	4352
113-116	4373
117-120	4182
121-124	4085
125-128	4426
129-132	4415
133-136	4180

Specimens have been molded for impact strength evaluation.

ADHESIVES Two new formulations (AD160 and AD161) of epoxy and epoxy-polyol modifications of MOCA*-polyurethane systems were prepared and cast into flexible films. Test specimens were cut after the films had been aged a minimum of seven days. Table 2 lists composition and stress-strain data for all formulations tested to date.

* Trademark of E. I. duPont de Nemours

Table 2

Test No.	Adiprene		MOCA ^a	AGE ^b	Epoxide			RD-2 ^f	Epon 8129	HET-Anhyd. ^h	% Elong.	T.S.* psi
AD-	L100	L167			201 ^c	206 ^d	269 ^e					
1	12	-	1.1	2.0	-	-	-	-	-	-	441	1687
3	12	-	1.1	-	1.1	-	-	-	-	-	678	2562
5	12	-	1.1	0.3	-	0.5	-	-	-	-	627	3634
102	21.6	-	1.94	-	-	-	-	-	1.56	-	763	3154
104	-	22.37	1.94	-	-	0.25	-	-	1.65	0.27	760	2646
105	-	21.6	1.94	-	-	0.25	-	-	2.34	0.27	820	1516
107	-	21.6	-	-	-	1.51	-	-	1.56	1.61	1163	1415
138	-	5.5	1.1	2.0	-	-	-	-	-	-	487	2941
139	-	5.5	1.1	-	1.1	-	-	-	-	-	568	3251
140	-	5.5	1.1	0.3	-	0.5	-	-	-	-	442	4536
141	12	-	2.49	2.0	-	-	-	-	-	-	106	543
142	-	5.5	2.27	2.0	-	-	-	-	-	-	126	467
143	12	-	1.91	-	1.1	-	-	-	-	-	223	670
144	-	5.5	1.69	-	1.1	-	-	-	-	-	44	272
145	12	-	1.95	0.3	-	0.5	-	-	-	-	201	536
146	-	5.5	1.73	0.3	-	0.5	-	-	-	-	88	538
147	-	5.5	2.0	-	-	1.0	-	-	-	-	58	662
148	-	5.5	1.9	-	-	-	1.0	-	-	-	44	539
149	-	5.5	2.0	-	-	0.5	0.5	-	-	-	50	666
150	-	5.5	1.8	-	-	-	-	1.0	-	-	98	466
151	-	5.5	1.8	-	-	-	-	-	1.0	-	406	1591
152	12	-	1.1	-	-	-	-	-	-	-	503	2265
153	10	-	1.1	-	-	-	-	-	-	-	560	1830
154	-	5.5	1.1	-	-	-	-	-	†	-	450	3007
155	-	5.5	1.1	-	-	1.0	-	-	-	-	500	3027
156	-	5.5	1.1	-	-	-	1.0	-	-	-	540	3688
157	-	5.5	1.1	-	-	0.5	0.5	-	-	-	433	1587
158	-	5.5	1.1	-	-	-	-	1.0	-	-	78	889
159	-	5.5	1.1	-	-	-	-	-	1.0	-	241	1080
160	-	5.5	1.1	-	-	0.5	1.0	-	-	-	174	533
161 [†]	-	5.5	1.1	-	-	1.0	-	-	-	-	138	395

Formulations are by weight

(1) 1 gram polyglycol P-400 added

(a) 4, 4' methylene-bis-(2-chloroaniline)

(b) Allyl glycidyl ether

(c) (3,4-epoxy-6-methylcyclohexylmethyl-3, 4-epoxy-6-methylcyclohexanecarboxylate) Union Carbide Chemicals Corporation

(d) Vinylcyclohexenedioxide - Union Carbide Chemicals Corporation

(e) Dipentene dioxide - Union Carbide Chemicals Corporation

(f) Butanediol diglycidyl ether

(g) Aliphatic epoxy resin - Shell Chemical Corporation

(h) 1, 4, 5, 6, 7, 7-hexachlorobicyclo-[2.2.1] -5 heptene-2, 3-dicarboxylic anhydride

* Tensile Strength

Formulations AD-1, 3, 5, 102, 104, 105, 107, 138-146 were included for the purpose of comparison and continuity. Formulations AD-141-151 show the effect of the addition of MOCA on a stoichiometric basis to match the addition of epoxy viscosity modifiers. AGE is a monoepoxide while Epoxide 201, 206, 269, RD-2, and Epon 812 are di- or polyepoxides. With the exception of Epon 812, their structures are such that the inhibited amine, MOCA, has low reactivity with them at room temperature. The addition of excess MOCA as in AD-141-151 causes incomplete cure as is shown by the tensile data. When it became apparent that excess MOCA hindered the cure of cast films, the formulation series AD-152 through AD-159 was made at the theoretical stoichiometric ratio of Adiprene L-167 to MOCA with the addition of the respective epoxy viscosity modifiers. Formulation AD-156 has excellent physical properties. Epon 812 is an aliphatic epoxy formed by reacting epichlorohydrin and glycerine, and resembles the bisphenol A-Epichlorohydrin epoxy systems. MOCA will cure Epon 812 slowly. A comparison of AD-151 and AD-159 shows that the addition of 0.7 gram MOCA as the stoichiometric equivalent of 1.0 gram Epon 812 in AD-151 produces a tensile strength of 1591 psi as compared to 1080 psi. Formulations AD-160-161 represent attempts to produce adhesives with better brushing characteristics.

The addition of 0.5 gram Epoxide 269 in AD-160 reduces the tensile strength from 1587 psi (AD-157) to 533 psi (AD-160). This weakening of the cast film indicates that the added Epoxide 269 did not enter completely into the curing reaction. A similar occurrence is noticed with the addition of 1.0 gram Polyglycol P-400 in AD-161 to the formulation AD-155. The tensile strength is reduced from 3027 psi to 395 psi.

There were variations in film thickness, porosity, and the degree of cure between the films and this factor should be considered when comparing tensile data.

ADHESIVE EXUDATE The study is continuing on the material exuded from a production item.

The absence of ionic nitrate in aqueous extract of the exudate was substantiated by a negative response to N, N'-diphenylbenzidine-concentrated sulfuric acid reagent, which is capable of detecting nitrate ions in a drop of 0.001 normal nitric acid solution.

In the continuing study of the stability of Versamid* 140 resin to exposure to the atmosphere a gallon can (lot 1A539) was opened and a fresh, clear solution in chloroform was made (10g/100 ml). The resin, however, is apparently sensitive to atmospheric moisture, since the surface became cloudy on continued exposure. Within half an hour the solution became turbid.

Based on the three lots (OG505, OBR506 and 1A539) tested, variation in color and color intensity of Versamid 140 was attributed to differences between lots and not to degree of aging and exposure. Visual observations were substantiated by spectrophotometric scanning.

Three separate samplings of the colored exudated material have been examined by infrared spectrometry. The material has been found by fractionation with solvents of different polarity to consist of more than one compound. In an effort to identify these materials 29 different samples of substances believed related to the materials have been studied. Infrared spectra of samples of fresh uncured epoxy, cured epoxy and Versamid-140 were made. Only Versamid-140 showed a spectra similar to that of the exudate. A double peak due to C-& stretching frequencies at 3.4 and 3.5 microns are similar in both spectra. The spectra are also similar at 5.95 and 6.8 microns. An infrared study was made on the effects of aging and the effect of water and dilute hydrochloric and nitric acids on Versamid-140. On aging, Versamid-140 gives a spectrum which still more closely resembles that of the exudate. Peaks appearing at 2.9, 5.95 and 6.1 microns become similar in shape and relative intensity. Hydrolysis of Versamid-140 with dilute acid produces a material containing a carbonyl peak similar to that of the exudated material.

* Trademark of Flood Machinery and Chemical Corporation

When Versamid-140 has been treated with acid, a dark colored substance results. The spectrum of the water soluble fraction of the exudate has what appears to be ionic nitrate. The conclusion is that the exudate appears to be Versamid-140 hydrolyzed by nitrate.

RADIOELEMENTS

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

IONIUM PROJECT Samples of ionium-bearing raffinates and residues were sent to Oak Ridge National Laboratory for neutron activation analysis of thorium-232. The results reported previously (MLM-CF-61-4-130, page 89) have been revised and are reported below:

THORIUM CONCENTRATIONS OF IONIUM-BEARING SAMPLES

Sample	Thorium-232 μ g	Thorium-230 (ionium) (μ g)	Ratio $\text{Th}^{232}/\text{Th}^{230}$
2A	2.79; 2.81 per ml.	0.031 per ml.	90
8D	1.12; 1.16 per ml.	0.020 per ml.	57
12A	0.86; 0.96 per ml.	0.016 per ml.	57
14C	2.30; 2.18 per ml.	0.0084 per ml.	267
15C	1.30; 1.18 per ml.	0.0087 per ml.	143
18E	95.0; 105; 96.7; 106 per g.	3.3	31
24AR	9.99 $\times 10^3$; 10.7 $\times 10^3$; 9.83 $\times 10^3$; 10.6 $\times 10^3$ per g.	1.1 per g.	$> 10^4$
24C	482; 505; 481; 505 per g.	7.0 per g.	70
28A	31; 26; 33; 26 per g.	3.8 per g.	7.6

The results show that the Mallinckrodt "airport cake" (Sample 28A) is the best source of ionium from the standpoint of isotopic ratio. Although the "Q-11 Metal oxides" (Sample 24C) from the Fernald, Ohio plant of National Lead Co. is richer in ionium, its isotopic ratio is poorer by an order of magnitude. In addition, most of the weight of the "airport cake" is due to lime added for neutralization, while the weight of the "metal oxides" represents the non-volatile calcined residue of the acid raffinate from solvent extraction. Thus, the "airport cake" would be easier to process for ionium.

HALF-LIFE OF RADIUM-223 The heat output of a five-millicurie source of radium-223 in equilibrium with its decay products is being followed by the Calorimetry Group. A sample of the purified radium-223 was set aside at the time of separation and has now been analyzed for actinium-227 by the lanthanum phosphate procedure (MLM-1108, page 9).

It was found that the product contained 0.103 microcuries (0.002 per cent) of the original actinium-227. The radium-223 yield was estimated to be 88 per cent. No thorium-227 was detected other than that which was attributable to growth from actinium-227 during the time between separation of the radium-223 and its analysis.

The small amount of actinium-227 found in the radium-223 sample is insufficient to account for the long-lived impurity detected by calorimetry. If radium-226 is present, it would not be found by the lanthanum phosphate analytical procedure, and its presence has not yet been detected by gamma spectroscopy.

PROTACTINIUM PROJECT Approximately 240 milligrams of protactinium-231 was recovered from raffinates, residues and analytical samples resulting from the FY 1956 protactinium project. The protactinium was concentrated in approximately 140 milliliters of 50 per cent diisobutyl carbinol in benzene (DIBC-bz).

After eight months, there was no indication of hydrolysis in the DIBC-bz solution. A small aqueous phase (approximately 1.8 milliliters) was removed and examined by gamma spectroscopy. It was found to contain actinium-227 and its decay products, but only a small amount of protactinium. The aqueous phase was made nine normal in sulfuric acid and six normal in hydrochloric acid and extracted with DIBC-bz. Approximately one milligram of protactinium-231 was recovered, and this was combined with the major organic fraction.

The organic solution was sampled, and an aliquot of the sample was alpha-counted. The total protactinium-231 was estimated to be 243 milligrams.

Niobium-95 was extracted with a portion of the DIBC-bz, and the organic fractions were recombined and intimately mixed. The mixed solution was sampled and gamma-counted, and it was found that the counting rate at 300 kev, representing protactinium-231, was approximately four times as great as that at 764 kev., representing niobium-95. This sample was retained as a gamma-counting standard. A sample of pure niobium-95 and a sample of pure protactinium-231 were also retained for use in determining the relative contribution of each to the 300 and 764 kev gamma counting levels. (Under the conditions used, niobium-95 contributes, by Compton effect, approximately 0.4 of its peak counting rate to the 300 kev level. Pure protactinium-231 contributes no significant count to the 764 kev level, although an aged sample of protactinium-231 may make a small contribution, due to the 764 kev levels of lead-211.)

Since spectrographic analysis was still not available, it was necessary to proceed with the purification of protactinium under the assumption that the only significant impurity was niobium and that, after separation of the protactinium, the residue on ignition would be niobium pentoxide. Accordingly, a sample of the DIBC-bz solution was stripped of protactinium and niobium, and the protactinium was separated by hydrolysis from a mixture of hydrochloric and oxalic acids.

The protactinium was centrifuged, and the supernatant liquid was set aside for several days. A small precipitate which settled out was filtered off, and the filtrate was turned over to the analytical group for treatment as follows:

To the solution containing niobium and a trace of protactinium, add an equal volume of concentrated hydrochloric acid and evaporate to a low volume. (Some niobium hydrolyzes and carries the residual colloidal protactinium.) Cool, filter through Whatman No. 42 filter paper, and wash with six normal hydrochloric acid. Evaporate the filtrate to dryness in a porcelain crucible, and ignite the residue at approximately 650 degrees centigrade. Transfer the residue to a tared vial and weigh.

The off-white residue was assumed to be niobium pentoxide. On the basis of this assumption and the gamma counting rate at 764 kev, it was found that the 243 milligrams of protactinium was contaminated with 219 milligrams of niobium (a ratio of 1.11).

Purification of the protactinium was carried out in batches containing 40 to 50 milligrams of protactinium. In a typical separation, each of three 10-milliliter portions of the DIBC-bz solution was stripped by stirring for five minutes with four milliliters of 10 per cent oxalic acid solution. The organic phase was sampled for gamma-counting and discarded. Each aqueous phase was scrubbed with four milliliters of benzene. The benzene was separated, gamma-counted, and discarded.

The aqueous phases, each of which contained a precipitate, was made to 10 milliliters with six normal hydrochloric acid, stirred five minutes and centrifuged. All three aqueous supernates were decanted to a single centrifuge tube, mixed, and sampled for gamma counting. To each precipitate, four milliliters of 10 per cent oxalic acid was added, and the mixture was stirred until the precipitate dissolved completely.

The oxalic acid solutions of protactinium were combined, and 12 milliliters of concentrated hydrochloric acid was added. A precipitate developed immediately. The mixture was stirred for approximately 30 minutes and centrifuged. The supernate was decanted and sampled for gamma-counting.

The precipitate was transferred to a beaker and digested with approximately five milliliters of seven normal sulfuric acid. Gas evolution indicated that oxalic acid was being decomposed. The temperature was raised slowly until all the oxalic acid had been destroyed, and fumes of sulfur trioxide were evolved. The mixture was allowed to cool to room temperature.

During the sulfuric acid digestion, the protactinium precipitate dissolved completely, but as the volume of the solution decreased, and the sulfuric acid concentration increased, a crystalline precipitate developed. When the mixture had cooled, the slurry was centrifuged and the sulfuric acid supernate was separated. Gamma-counting showed that the residual niobium concentrated in the sulfuric acid solution, leaving essentially pure protactinium crystals (probably $\text{Pa}_2(\text{SO}_4)_3 \cdot x \text{H}_2\text{O}$).

The sulfuric acid solution, which contained relatively little protactinium, was used repeatedly for the final digestion of the various batches. When all of the protactinium had been recovered from the DIBC-bz and purified, the saturated sulfuric acid solution was diluted, made six normal in hydrochloric acid, and extracted with fresh DIBC-bz to recover the protactinium (approximately 4.6 milligrams in two milliliters of concentrated sulfuric acid).

All aqueous and organic solutions containing more than two micrograms of protactinium per milliliter were reprocessed, and the protactinium was combined and purified. The final product had a purification factor of over 800, based on the gamma counting rates at 300 and 764 kev. Essentially all of the protactinium in the DIBC-bz is accounted for.

A final assay by alpha-counting has not yet been made, because it is now necessary to repurify and combine various samples taken during processing in FY 1957 and 1958. It is estimated that an additional 100 milligrams of protactinium will be recovered.

The results of the oxalic acid purification procedure are given in Table 3. The results of the sulfuric acid digestion are shown in Table 4. It should be noted that the results in the two tables cannot be directly related to each other, inasmuch as the same sulfuric acid solution was used repeatedly, and the niobium present was an accumulation from several such digestions.

The purified protactinium is being stored in five normal sulfuric acid solutions at a protactinium concentration of approximately 30 milligrams per milliliter.

Table 3

SEPARATION OF PROTACTINIUM FROM NIOBIUM BY PRECIPITATION
FROM HYDROCHLORIC-OXALIC ACID

(Total Protactinium = 56 mg.; Total Niobium = 51 mg.)

Fraction	Per Cent Protactinium	Per Cent Niobium
Stripped organic	0.2	1.9
Benzene scrub	0.001	0.01
First aqueous supernate	3.1	94.8
Second aqueous supernate	1.1	3.9
Total	4.4	100.6

Table 4

SEPARATION OF PROTACTINIUM FROM NIOBIUM BY CRYSTALLIZATION
FROM CONCENTRATED SULFURIC ACID

(Total Protactinium = 76.1 mg.; Total Niobium = 0.24 mg.)

Fraction	Per Cent Protactinium	Per Cent Niobium
H ₂ SO ₄ solution	9.5	85.3
Solid Phase	<u>90.5</u>	<u>14.7</u>
Total	100.0	100.0

ISOTOPE SEPARATION AND PURIFICATION

Processes are being developed for separating and purifying the isotopes of a number of elements including hydrogen, the noble gases, carbon and uranium. Potential sources of supply of these materials are being evaluated.

URANIUM SEPARATION Gas centrifugation and photochemical separation are particularly applicable to separating research quantities of uranium isotopes. Since gas centrifugation has been relatively well developed, the method offers the desirable production capacity with a reasonable amount of development. Although the photochemical method will require considerable research and development, it appears to offer a highly suitable method for removing trace contaminants from relatively pure isotopes.

As a result of the preliminary investigation, described in last month's report, the following studies were begun:

Gas Centrifugation The available literature on this subject was studied to become better acquainted with the theory and with the operating parameters. Gas centrifugation is particularly attractive for separating heavy element isotopes because the separation of binary mixtures is a function of the mass difference only. For most isotope separation processes, the separation is a function of the relative mass difference.

The development of the countercurrent flow system has made the method feasible for a relatively large throughput. This system combines the centrifugal effect with a gas convection flow in a direction parallel to the axis of the centrifuge. This arrangement cascades the centrifugal separation throughout the axial length of the centrifuge bowl. The "long bowl" gas centrifuges with the countercurrent flow feature have separated uranium-235 from uranium-238 with a separation factor as high as 1.4.

Two schemes have been developed to generate the countercurrent gas flow. In one method the convection current is driven thermally, and in the other method special scoops are inserted in the gas stream at appropriate locations in the centrifuge bowl.

G. Zippe, ORO-315, "Development of the Short Bowl Ultracentrifuge," July, 1960

The scoops serve a dual purpose in driving the countercurrent flow and in building the effluent gas pressure from the centrifuge to a more practical value.

W. Groth, et. al AEC-TR-4264 "Separation Potential of Thermally Driven Countercurrent Gas Centrifuges," August, 1960

An investigation of the relative merits of these two schemes was started. It is apparent that much more experimental work must be done to optimize this countercurrent flow to design a feasible uranium isotope purification plant.

Preliminary work on this system will use xenon or krypton.

Photochemical Separation A review of the literature indicates that only a small effort has been made to initiate reactions of photoexcited uranium. Considerable work with photoexcited isotopes of mercury has been done, however, by Gunning, et. al. Gunning's work is being studied to gain some insight into possible photocatalysis of reactions with uranium.

H. E. Gunning, et. al, Can. J. Chem. 37, 1432 (1959)

Because of the extreme complexity of the uranium spectrum and because of the uncertainty of the photochemical process in uranium, considerable experimental work will be required to determine if a satisfactory process can be developed.

In order to investigate the photochemical method in view of isotope separation, a suitable narrow-band radiation source is required which can be tuned to some strong line in the uranium absorption spectrum

which exhibits a good isotopic shift. Some efforts have been made to locate a wide-aperture monochrometer which can be used for low intensity irradiation studies. However, it is doubtful that sufficient intensity can be obtained from a monochrometer to catalyze large-scale reactions unless a suitable high intensity light source of more or less monochromatic radiation can be developed.

Recent successes with optical masers have made this type of device very attractive. The beam from a maser is very intense and extremely monochromatic. However, masers available at present are highly specialized as to output frequency and the variety of frequencies available is quite limited. A small amount of thermal tuning of a solid state maser is possible.

A ruby maser with a beam of 6943A was purchased for preliminary studies of photochemical reactions of UF_6 in various media.

XENON PURIFICATION AND ISOTOPE SEPARATION

Gas Chromatography The experimental gas chromatograph resolved the krypton-xenon peaks and effected an overall decontamination factor of 15.

Low Temperature Distillation The analytical system is being calibrated by running duplicate samples in the ion chamber and the beta counting system. The effect of total pressure in the ion chamber has been found to be not too critical. Adjustment of the pressure to ± 1 mm. Hg has negligible effect.

Several runs have been made on the still to find the proper resistances for automatic pressure control using the condenser and, hence, the liquid nitrogen boil-off as a control. Runs will be possible at constant still-pot heat and, hence, constant vapor velocity.

Analytical The proportional counting system has been calibrated volumetrically by gas expansion. Plateau and background runs have been made on a one-liter proportional counter.

Thermal Diffusion The draw off systems in the present location have been more dependable and greater than 99 per cent neon-22 is issuing from a two-stage separation. Neon is used as a checkout gas on the flowing system due to its lower cost.

HELIUM ISOTOPE SEPARATION AND PURIFICATION

Thermal Diffusion Since installation of the wire temperature balancing resistors and replacement of the solenoid valves, the feed flow rate has been nearly doubled. The design needed for reinstallation of this system has been nearly completed.

CARBON ISOTOPE SEPARATION Theoretical evaluations of carbon-13 separation by thermal diffusion are being conducted using the experimental data obtained from the five-column series system. The interim period during construction will be spent studying the thermal stability of carbon monoxide and methane, circulation systems for cascade loops and evaluation of carbon monoxide to methane conversion for optimum process variables.

KRYPTON SAMPLE PREPARATION After introducing enriched feed into the five-column series system, the krypton-78 and krypton-80 content rose to around 55 per cent. These columns have now been shut down due to the carbon-13 construction. The product obtained from this operation will be further enriched in the two column cascade system and then preferential enrichment of krypton-78 and krypton-80 should follow naturally. The object is to obtain two samples which are enriched to at least 20 per cent in krypton-78 and krypton-80. One sample should have the 78/80 ratio equal to ten, the other the 80/78 ratio equal to ten.

REACTOR FUELS AND MATERIALS DEVELOPMENT

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

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

Density The density of liquid lanthanum was determined from 940° to 1000°C by the vacuum pycnometer method. The results are shown in Table 5 and Figure 1. Also shown in Figure 1 are four values determined at Ames Laboratory (ISC-659) in which the sessile-drop method was used.

Table 5

DENSITY OF LIQUID LANTHANUM

Temperature, °C	Density, g/cc.
945	5.955
948	5.942
978	5.940
1000	5.926

The thermal coefficient of cubical expansion for solid lanthanum is 24×10^{-6} cc/cc°C. With the assumption of a constant expansion coefficient to its melting point, 924°C, the density of solid lanthanum would be 6.06 g/cc. while the density of liquid lanthanum at 924°C, as found by extrapolation in Figure 1, is 5.96 g/cc. The volume expansion upon fusion is, therefore, about 1.7 per cent.

The tantalum pycnometers with a volume of slightly over 0.5 cc. were calibrated at 28.85°C with mercury to an accuracy of approximately ± 0.0005 cc. A volume expansion coefficient of 19×10^{-6} cc/cc°C was used to correct for the volumes of the pycnometers at test temperatures.

Viscosity Investigations into the fundamental characteristics of the oscillating cup viscometer have continued with a determination of the behavior of 0.010-inch diameter torsion fibers. The viscosities of lanthanum and bismuth metals were checked with both a 0.010-inch diameter platinum-tungsten alloy fiber and a molybdenum fiber.

Preliminary evaluation indicated that both of the stiffer fibers were giving viscosity values 10 to 30 per cent higher than those obtained with the 0.005-inch diameter fibers.

Since the period of oscillation had changed from 12 seconds to three seconds when the stiffer fibers were substituted, the system would have had a much greater angular velocity if a constant amplitude of oscillation were used with both fibers. The initial amplitudes of the oscillations were decreased, therefore, from 38° to 13° to decrease the angular velocity.

A determination was made with molten bismuth at 524°C to determine if the logarithmic decrement was independent of amplitude with the stiff fiber. When amplitudes of 6°, 13° and 19° were used, the logarithmic

DENSITY OF LIQUID LANTHANUM

Figure 1

decrement was found to increase in value with the amplitude. This observation indicates that the spurious viscosities observed with the stiffer fibers for lanthanum and bismuth resulted from non-laminar flow encountered because of the high angular velocity of the crucible.

High Temperature Calorimeter Construction of the calorimeter is now complete and the assembly is being calibrated at room temperature.

The zero unbalance of the calorimeter bridge is approximately 2278 microvolts at 10 milliamperes bridge current. This unbalance is greater than desirable but it can be corrected by either an adjustment in the platinum bridge arms or by the addition of a platinum resistance in series with the output of the bridge together with the application of a "bucking" voltage across it. Each method has its advantages. In its present configuration the zero unbalance must be subtracted from each reading. The unbalance will be corrected later when the calorimeter is ready to be tested at higher temperatures.

Meanwhile, much information has been obtained from the calorimeter, and it appears to be functioning well within the expected accuracy. A polonium-210 sample, whose actual output was 1.0604 watts, was placed into the sample side of the calorimeter and allowed to reach equilibrium. After the sample was removed, current was supplied to the heater to duplicate the output of the sample. Calculations showed that the calorimeter had measured an output from the sample of 1.0566 watts, within 0.36 per cent of the actual value.

The physical properties of the calorimeter system which have been determined are listed below:

Property	Value	Units
Sensitivity (Dummy Side)	3112.9	microvolts/watts
Sensitivity (Sample Side)	2547.03	microvolts/watts
Half Life $T_{1/2}$	200.11	seconds
Time to reach 0.01 per cent of equilibrium	44.3	minutes
Thermal Resistivity $T_{th.}$	2.1744	$^{\circ}\text{C}/\text{watt}$
Heat Capacity $^{\circ}\text{C}$	129.782	joules/ $^{\circ}\text{C}$
Time Constant $T_{const.} = \frac{1}{RC}$	3.54×10^{-3}	seconds $^{-1}$
$T_{const.}^{-1} = RC$	282.198	seconds

The heat distribution error of the sample side is presently being determined, with a small heat source placed at various positions in the sample cavity.

The nickel block will next be placed under controlled heat. This change will enable the block to reach equilibrium state faster and will serve as a test of the efficiency of the controller bridge and heater.

Metallography An investigation was conducted into the compatibility of Haynes 25 cobalt alloy alloy with plutonium at 900 $^{\circ}\text{C}$. A 0.136-inch hole was drilled in a small block of the Haynes 25 alloy, and a small piece of plutonium was placed in the hole. The alloy was then heated under vacuum to about 900 $^{\circ}\text{C}$ and held at this temperature. Accurate temperature measurement was not possible because a fogged viewing port obstructed the sighting with an optical pyrometer. After one hour the sample was removed and polished metallographically. Penetration measurements were made, and photomicrographs were taken of the sample at various magnifications.

The sample was then placed back in the vacuum system and again held at about 900 $^{\circ}\text{C}$ for an additional three hours and then metallographically polished and re-examined.

Measurements on the average diameter of the hole after one hour treatment indicated an increase of 0.0175 inch on the radius. After three additional hours at 900°C, the average radial increase was 0.0178 inches indicating that very little penetration took place after the first hour of treatment since solid state equilibrium had apparently been reached between the alloy and the plutonium so that solid state diffusion became the controlling factor.

PLUTONIUM BEARING GLASS FIBERS *At the request of the Reactor Fuels and Materials Development branch of the AEC, a cooperative experiment in the fabrication of plutonium-bearing glass fibers is being carried out by Rensselaer Polytechnic Institute, Owens-Corning Fiberglas Corporation and Mound Laboratory.*

Six samples of plutonium bearing glass fibers were encapsulated in quartz tubes and sealed in aluminum cans for irradiation studies in the Brookhaven National Laboratory reactor. The sealed bulbs contain about three atmospheres pressure of helium at room temperature. The parent glass was 70 weight per cent silicon dioxide and 30 weight per cent sodium monoxide. Strands were cut from a coil of fibers and were weighed before encapsulation. Each of the samples weighed approximately 50 milligrams and contained fifteen weight per cent plutonium oxide.

These fibers were approximately 15 microns in diameter and exhibited an average tensile strength of 101,500 pounds per square inch.

An irradiation time schedule has been set up for removal of one sample from each group after 1, 2, 4, 19 and 38 days.

The samples scheduled for 1, 2, 4, and 8 days will experience a flux of 0.95×10^{13} n/sec/cm² and temperatures of 50°-70°C. Those scheduled for 19 and 38 days will experience a flux of 0.7×10^{13} n/sec/cm² and a temperature of 70°C.

To determine the efficiency of escape of alpha particles from fibers of plutonium-bearing glass, a one-inch long fiber containing 10 weight per cent plutonium oxide and having a diameter of 28.3 microns was mounted in a holder which provided complete exposure of the fiber. Alpha disintegrations were counted in an ionizing gas counter. The results showed 360,000 disintegrations per minute. A theoretical calculation was made assuming the density of the glass fiber to be 2.5 g/cc. The density of the massive 10 weight per cent plutonium oxide glass is probably closer to 3 g/cc. but the fiber is quenched in air and still retains the density of the glass at a higher temperature. The calculated value, assuming 2.5 g/cc. is 500,000 disintegrations per minute. Based on this value, approximately 72 per cent of the alpha particles escaped from the fiber.

Further assays are being made on fibers taken from a single long length to determine the uniformity of distribution of plutonium oxide in the glass fibers.

Eight new glass compositions received from the Owens-Corning Fiberglas Corporation have been evaluated for their ability to dissolve 15 weight per cent plutonium oxide. This composition was selected for evaluation since a very durable glass of this composition has been prepared. The compositions of these glasses are listed in Table 6.

Glasses X-1227 was the most promising for ease of dissolution of plutonium oxide. This composition readily dissolved 15 weight per cent plutonium oxide after only 15 minutes at 1650°C. An attempt will be made to incorporate 20 weight per cent plutonium oxide in this glass and its resistance to water and dilute acids will be determined.

Table 6

**COMPOSITIONS OF EIGHT PARENT OWENS-CORNING GLASSES WHICH WERE EVALUATED
FOR THEIR ABILITY TO DISSOLVE 15 WEIGHT PER CENT PLUTONIUM OXIDE AT 1650°C**

No.	SiO ₂	Al ₂ O ₃	CaO	MgO	BaO	Na ₂ O	K ₂ O	F ₂	Fe ₂ O ₃	Remarks and Cation Per Cent Pu
X-1225	64.97	5.41	5.41	-	-	16.24	5.41	2.44	0.12	c/o = 3.27 15 weight per cent is the limit
X-1226	63.40	5.29	5.29	-	-	15.86	4.76	4.76	0.11	c/o = 3.39 15 weight per cent is the limit
X-1227	69.42	2.56	5.58	-	-	16.73	5.58	-	0.12	c/o = 3.24 Dissolved 15 weight per cent readily
X-1228	63.75	9.57	5.31	-	-	15.94	5.31	-	0.12	c/o = 3.2 would not dissolve 15 weight per cent
X-1229	64.08	5.55	5.55	2.50	-	16.65	5.55	-	0.12	c/o = 3.17 would not dissolve 15 weight per cent
X-1230	66.58	5.55	4.00	1.55	-	11.65	10.55	-	0.12	c/o = 4.44 marginal
X-1231	66.58	5.55	4.00	1.55	-	20.20	2.00	-	0.12	c/o = 3.18 Dissolved 15 weight per cent readily
X-1232	65.59	5.47	3.94	1.53	1.48	16.40	5.47	-	0.12	c/o = 3.14 would not dissolve 15 weight per cent

The high alumina content in X-1228, present at the expense of silicon dioxide, lowered the ability of the glass to dissolve plutonium oxide. After 75 minutes at 1650°C this glass had dissolved very little plutonium oxide. The alumina apparently weakens the silicon dioxide network so that devitrification occurred.

Glass X-1229 would not dissolve plutonium oxide to the extent of 15 weight per cent.

Compositions X-1230 and X-1231 are identical except for their relative contents of sodium oxide and potassium oxide. The combined weight per cent of the two were the same in each glass. X-1231 dissolved 15 weight per cent plutonium oxide completely and without difficulty. X-1230 would not dissolve 15 weight per cent completely. This effect is probably due to the larger size of the potassium cation.

X-1232 would not dissolve 15 weight per cent after 75 minutes at 1650°C.

The most promising glasses, X-1231 and X-1227, will be evaluated for their ability to dissolve 20 weight per cent plutonium oxide. If successful, the chemical durability of these glasses will be investigated.

Glass Neutron Standard A glass neutron standard was prepared which had a neutron flux of 3000 neutrons per second for a six gram sample. It contained 0.8310 grams of plutonium oxide and 0.4980 grams of beryllium oxide which is a ratio of 6.50 beryllium atoms to one plutonium atom.

The parent glass used for this standard was a glass containing 70 weight per cent silicon dioxide and 30 weight per cent sodium oxide. This glass is hygroscopic and remains so when plutonium oxide is added. However, the neutron standard does not exhibit a hygroscopic tendency.