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# MOUND LABORATORY MONTHLY PROGRESS REPORT FOR FEBRUARY, 1961

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

These reports are not intended to constitute publication in any sense of the word. Final results either will be submitted for publication in regular professional journals or will be published in the form of MLM topical reports.

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# MONSANTO RESEARCH CORPORATION

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# MOUND LABORATORY

MIAMISBURG, OHIO

UBSIDIARY

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# UNITED STATES ATOMIC ENERGY COMMISSION

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1) Total (p. 2. c. )
 <li1) Total (p. 2. c. )</li>

#### 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

One experimental diallyl phthalate formulation was made:

|                                     | Parts Per 100 Parts Resin                           |
|-------------------------------------|---|
|                                     | (Dapon <sup>1</sup> -35 + Oxiron <sup>2</sup> 2002) |
| Kaomica <sup>3</sup>                | 40  |
| C-3000 English Mica <sup>4</sup>    | 60  |
| Hydroquinone                        | 0.03  |
| Harshaw Scarlet <sup>s</sup> , 25AD | 1.0   |
| Calcium Stearate                    | 2.0   |
| t-Butyl Perbenzoate                 | 2.0   |
| Phthalic Anhydride                  | 0.91  |
| 1-4-Butanediol                      | 0.2   |
| Acetone                             | 200   |
|                                     |   |

Processing was identical with that used with standard DAP formulations with the exception that the material was not placed in the sigma-blade mixer after high speed stirring, but was placed directly on the differential roll mills.

Cups molded from this compound were rubbery and not fully cured after a four minute press cycle (standard for mineral filled DAP formulations). A cure of eight minutes was adequate. Molded surfaces looked good and the cup was hard. The slow cure was probably the result of low diol and/or anhydride content. Only 0.15 mole of anhydride was used per mole of epoxy.

#### ADHESIVES

A survey and re-evaluation of all previous work on polyurethane and polyurethane-epoxy copolymer systems has been initiated to determine which phases are more productive than others, and which have been neglected or touched upon only lightly.

Nineteen additional formulations involving both epoxy and polyol-modified polyurethane systems are being reported this month. All weights are given in grams.

- <sup>1</sup> Trademark of Food Machinery and Chemical Corp.
- <sup>2</sup> Trademark of Food Machinery and Chemical Corp.
- <sup>3</sup> Trademark of Edgar Plastics and Kaolin Co.
- <sup>4</sup> Trademark of the English Mica Co.
- <sup>5</sup> Trademark of Harshaw Chemical Corp.

| Formulation           | AD113   | AD114     | AD115    | AD116      | AD117   | AD118 | AD119   |
|-----------------------|---------|-----------|----------|------------|---------|-------|---------|
| ADL-213               | 7.76    | 7.76      | 7.76     | 7.76       | 7.76    | 7.76  | 7.76    |
| Moca²                 | 2.80    | 2.80      | 2.80     | 2.80       | 2.80    | 2.80  | 2.80    |
| Epon 812 <sup>3</sup> | 1.56    | 1.56      | 1.56     | 1.56       | 1.56    | 1.56  | 1.56    |
| 1,4 Butanediol        | 1.0     | 2.0       | 3.5      | 3.0        | 4.0     | 6.0   | 8.0     |
| Pot life Hrs.         | Too     | Тоо       | Too      | Too        | Τοο     | Ж     | 4       |
|                       | viscous | viscous   | viscous  | viscous    | viscous |       |         |
|                       | none    | none      | none     | none       | none    |       |         |
| Time required         | Tough   | Tough     | So ft    | Non-       | Tacky   | Tacky | Very    |
| to cure to non-       | film    | film      | slightly | tacky      | jell'   | jeli  | viscous |
| tacky film            | low     | low Elon- | tacky    | film       | > 14    | > 14  | > 14    |
| (days)                | Elonga- | gation    | film     | low        |         |       |         |
|                       | tion-3  | tears     | > 14     | tear       |         |       |         |
|                       |         | easily    |          | resistance |         |       |         |
|                       |         | 3-1/2     |          | 7          |         |       |         |

# EFFECT OF POLYOL CONTENT ON EPOXY MODIFIED POLYURETHANES

<sup>1</sup> Adiprene-L213 Trademark of E. I. duPont de Nemours

2 4,4' methylene-bis-(2-chloroaniline) Trademark of É. I. duPont de Nemours

<sup>3</sup> Trademark of Shell Chemical Corp.

**A** 

The above series of formulations shows the effect of varying excess polyol content while fixing the Adiprene, MOCA catalyst, and epoxy contents. Also it was an attempt to measure the degree of reactivity of Adiprene L-213 as compared to Adiprenes L-100 and L-167 as reported in last month's report.

The data below compares the effect of several di-epoxide reactive diluents.

(י)

| Formulation           | AD120     | AD 121   | AD122   | AD123    |
|-----------------------|-----------|----------|---------|----------|
| ADL-213               | 7.76      | 7.76     | 7.76    | 7.76     |
| MOCA                  | 2.80      | 2.80     | 2.80    | 2.80     |
| 1,4 Butandiol         | 2.00      | 2.00     |         |          |
| UNox Epoxide 206(1)   | 0.82      |          | 4.      |          |
| Epoxol 7–4(²)         | ••        | 2.47     | 4.94    | ••       |
| R D-2( <sup>3</sup> ) | <b>`-</b> |          | ••      | 2.52     |
| Pot Life (Hrs.)       | Τοο       | 1/4      | 1       | 1/2      |
|                       | Viscous   |          |         |          |
|                       | (None)    |          | ×       |          |
| Cure Time (Days)      | 2         | 4        | · · 4   | 3        |
| Remark s              | Clear     | Cloudy   | Opaque  | Clear    |
|                       | Soft      | Soft     | White   | Elastic  |
|                       | Film      | Film     | Film    | Film     |
|                       | Poor      | Very     | Non-    | Medium   |
|                       | Tear      | Poor     | Elastic | Tear     |
|                       | Strength  | Tear     | Brittle | Strength |
|                       | -         | Strength | Tough   |          |

# THE EFFECT OF DI-EPOXIDE MODIFIERS ON POLYURETHANES

(1) Vinyl cyclohexane dioxide - Union Carbide Chemical Co.

(2) Polyepoxide derived from Soy Bean Oil - Swift & Company

(<sup>3</sup>) Butanediol diglycidyl ether, CIBA Products Corporation

Epoxol 7-4 was used to slow down the rate of cure of the system and reduce initial viscosity. The amounts used in formulations AD121 and 122 are (1) equal to and (2) twice the quantity equivalent to 1.56 grams Epon-812 used in earlier formulations. Evidently, this epoxy has fairly well defined limits of solubility in the polyurethane system. ÷ .

Until the present, very little work has been done to define the relative amounts of 1,3 butanediol, and 1,4 butanediol that the system Adiprene L-213 - ferric acetyl acetonate (FAA) can tolerate and still be thoroughly cured. The data below supplies this information.

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#### THE EFFECT OF 1,3 BUTANEDIOL ON ADL-213 - FAA SYSTEM

| Formulation     | AD124       | AD125            | AD 126       | AD127        |
|-----------------|-------------|------------------|--------------|--------------|
| ADL-213         | 25          | 25               | 25           | 25           |
| FAA             | 0.026       | 0.026            | 0.026        | 0.026        |
| 1,3 Butanediol  | 1.0         | ••               | 2.0          | 3.0          |
| 1,4 Butanediol  |             | 1.0              |              |              |
| Pot Life (Hrs.) | Non e-too   | None-too         | 2+           | 2+           |
|                 | viscous     | viscous          |              |              |
| Cure Time       |             |                  |              |              |
| (Days)          | 2           | 2                | 7            | 8            |
| Remarks         | Bubbles in  | Bubbles          | Clear tough  | Clear soft   |
|                 | film-other- | in film          | elastic film | elastic film |
|                 | wise clear  | otherwise        | good tear    | poor tear    |
|                 | tough elas- | tough clear      | resistance   | resistance   |
|                 | tic-good    | film-elas-       |              | Not thorough |
|                 | tear        | tic-good         |              | cure         |
|                 | resistance  | resistance       |              |              |
| Formulation     | AD 128      | AD 129           | AD130        | AD131        |
| ADL-213         | 25          | 25               | 25           | 25           |
| FAA             | 0.026       | 0.026            | 0.026        | 0.026        |
| 1,3 Butanediol  | 3.0         |                  | ° ==         |              |
| 1,4 Butanediol  | 1.0         | 4.0              | 6.0          | 7.0          |
| Pot Life (Hrs.) | 1/4         | Non <b>e-too</b> | None-too     | 1/6          |
|                 |             | viscous          | viscous      |              |
| Cure Time       |             |                  |              |              |
| (Days)          | > 10        | > 10             | No Cure      | No Cure      |
| Remarks         | Clear Soft  | Clear Soft       | Viscous      | Viscous      |
|                 | Film        | Film             | Liquid       | Liquid       |

1,3 butanediol has one primary and one secondary hydroxyl group while 1,4 butanediol has two primary hydroxyls and is much more reactive. This is shown in the preceding data as formulations AD126-AD131. Eight parts polyol per hundred parts resin gives a pot life of two hours when 1,3 butanediol is used. Sixteen parts polyol per hundred parts resin gives a formulation that has no pot life at all when 1,4 butanediol is used. The formulation thickens during degassing and cannot be applied as a film. 1%

Additional work is planned in formulating polyurethane systems in which different polyurethanes are compounded with epoxy modifiers to give a system which has properties intermediate between the two systems.

#### RADIOELEMENTS

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

**IONIUM PROJECT** A survey is being made of potential sources of kilogram quantities of ionium (thorium-230) in the United States. Preliminary examination of samples from uranium mills and processing plants has indicated several promising sources. These materials are being examined further.

For the preliminary analysis, solid samples were digested with hot concentrated nitric acid. While this relatively mild treatment solubilized only the readily available thorium, it was considered sufficient to establish the orders of magnitude of the ionium concentrations in the various samples.

Four of the solid samples were found to have ionium concentrations of one part per million or greater. These samples are being re-analyzed with a more vigorous digestion procedure.

Three samples of "Q-11 metal oxides" (# 24C) were treated with various reagents. The digestion solutions were collected and analyzed by the tributyl phosphate-cerium fluoride method reported previously (Interim Progress Report for January, 1961). The residues were gamma-counted at the 68-kev peak of ionium, and the total residual count was ascribed to ionium.

A single perchloric acid digestion did not remove all of the 68-kev gamma activity from the samples. The residues were treated as follows in an effort to solubilize all the radioactive material:

- 1. A 2.4 sample was digested with 10 ml of hot conc. HC10, containing some HNO, and HF. It was leached with HC1 and HNO, at room temperature, digested with 5 ml of hot 50 per cent NaOH solution, leached again with HC1 and HNO, and, finally, with 48% HF.
- 2. A 2.8 sample was leached twice with 20 ml of 3N HC1, digested with 10 ml of hot 70% HC10<sub>4</sub> containing HNO<sub>3</sub> and HF, leached with 20 ml of hot 15% NaOH, and digested again with 5 ml of hot 70% HC10<sub>4</sub>.
- 3. A 2.6 sample was leached twice with 20 ml of 3<u>N</u> HNO<sub>3</sub>, then digested to 10 ml of hot 70% HC10<sub>4</sub>.

Results of the analyses of the three composite solutions from these treatments are given in Table 1. The indicated ionium concentration (about seven parts per million) is nearly three times as great as that previously reported.

#### Table 1

# ANALYSIS OF Q-11 METAL OXIDE (#24C) FOR IONIUM (MICROGRAMS OF IONIUM PER GRAM OF SAMPLE)

|                    | <u> </u> | 2            | 3             |
|--------------------|----------|--------------|---------------|
| Digestion Solution | 7.1      | 6.9          | 7.0           |
| Residue (Maximum)  | 0.0      | <u>0.1</u> . | <u>N.A.</u> * |
| TOTALS             | 7.1      | 7.0          | <b>~</b> 7.0  |

\* N.A. - Not Available at report time.

Exploratory work has been done on the efficiency of hydrochloric, sulfuric and oxalic acids and of sodium hydroxide in recovering the ionium from the Q-11 Metal Oxides. In general, these reagents remove approximately one half of the gamma activity. It is not known, however, what proportion of the residual activity is due to ionium.

**HALF-LIFE OF RADIUM-223** The decay of a specially purified radium-223 sample has been followed by alpha counting for 83 days. Preliminary results for the first 50 days were reported last month. The half-life has been recalculated to include the new data, and the results are again reported as a function of the resolution time of the gas flow proportional counter (Table 2).

### Table 2

## HALF-LIFE OF RADIUM-223 AS A FUNCTION OF COUNTER RESOLVING TIME (T) (83 days - 31 observations)

| Т            | Half•Life | Probable Error |
|--------------|-----------|----------------|
| (Min. x 10') | (Days)    | (Days)         |
| 1.60         | 11.4004   | 0.0055         |
| 1.65         | 11.3808   | 0.0053         |
| 1.70         | 11.3602   | 0.0057         |

On the basis of the data obtained thus far, the most probable value of the resolution time of the counter is  $1.65 \times 10^{-7}$  minute. The most probable value of the half-life is  $11.3808 \pm 0.0053$  days.

A composite solution of actinium-227 in equilibrium with its decay products (AEM) was prepared from various ion-exchange tailings, actinium oxalate filtrates, and analytical samples taken during the FY-1952 processing of actinium. The solution was made approximately six normal in hydrochloric acid and passed through a small Dowex-1x8 anion-exchange column to remove iron. The column retained a substantial amount of radioactivity, principally thorium-227, as indicated by gamma spectrometry.

The hydrochloric acid effluent was evaporated to dryness and turned over to the Calorimetry Group for assay. The heat output of the sample was equivalent to that from 35.32 millicuries of polonium-210.

Because the five alpha emitters of the AEM group have an average alpha energy approximately 20 per cent higher than that of polonium-210, the equivalent actinium-227 content of the sample is 5.8 millicuries, assuming secular equilibrium. However, because of the loss of thorium-227 in processing, equilibrium is not assured, and the heat will be measured again after a lapse of several weeks.

The composite AEM sample is to be used for the recovery of several millicuries of radium-223. The decay of the radium-223 will be followed in one of the microcalorimeters and, subsequently, in a low geometry alpha counter.

The lanthanum phosphate procedure for determining actinium-227 and thorium-227 in purified radium-223 (Progress Report for December, 1960) has been checked with a standard AEM solution to determine the amount and nature of the losses on repeated precipitation. Instead of the supernates from each of three precipitations being discarded, they were analyzed separately for actinium-227 and thorium-227. The results (Table 3) indicate that the average loss per precipitation is less than one per cent.

#### Table 3

## LOSSES OF ACTINIUM-227 AND THORIUM-227 IN REPEATED PRECIPITATIONS OF LANTHANUM PHOSPHATE

| Fraction         | % Ac <sup>227</sup> | % Th²²ァ |
|------------------|---------------------|---------|
| Final LaPO4 ppt. | 97.6                | 97.3    |
| First Supernate  | 0.9                 | 0.7     |
| Second Supernate | 1.3                 | 0.5     |
| Third Supernate  | 0.7                 | 0.4     |
| Material Balance | 100.5               | 98.9    |

**DETERMINATION OF COINCIDENCE CORRECTION** A method is being developed for determining the coincidence correction in proportional alpha counters by following the decay of a short-lived radioisotope. The radioisotope being used for this purpose is lead-211, a 36-minute beta-emitter, which decays to bismuth-211 and polonium-211, two alpha-emitters having half-lives of 2.16 minutes and 0.52 second, respectively. The procedure for obtaining carrier-free lead-211 was described in last month's report.

To improve the precision of timing the counting interval, a switch clock and scale-of-1000 decade scaler have been modified to control the operation of the proportional alpha counters whose resolving times are being determined. A 60-cycle pulse generator feeds pulses to the timing scaler only when the proportional counter is operating. The input plate circuits of both scalers are opened or closed simultaneously by a single relay. The pulses recorded by the timing scaler can be applied as a correction factor for any inaccuracy in the operation of the mechanical switch clock.

The precision of radioactive counting, exclusive of human and instrument errors, is a function of the total number of counts recorded. The standard deviation is equal to the square root of the total number of counts recorded, regardless of the counting rate. Thus, it is possible to increase the precision of counting by increasing the time interval during which counts are recorded.

If the half-life of the radioisotope being counted is long compared to the counting interval, the effective time corresponding to the recorded count does not differ significantly from the midpoint of the counting interval. However, if the counting interval is a significant fraction of the half-life, the effective time of the average recorded counting rate may differ appreciably from the midpoint of the counting interval.

If a half-life is being determined by counting, it is necessary to evaluate any errors which may arise from an increase in the counting interval and to determine the extent to which the value of the half-life is affected by such an increase.

If the resolving time and the geometry of the counter are neglected, the average count recorded is the sum of all the radioactive events occurring during the counting interval:

$$\overline{R}' t = \int_{0}^{t} R' dt = R'_{0} \left[ \frac{1 - e^{-\lambda t}}{\lambda} \right] \qquad (1)$$

where t is the counting interval,  $\lambda$  is the radioactive decay constant,  $R'_0$  is the instantaneous counting rate at the beginning of the counting interval, and  $\overline{R'}$  is the average counting rate during the counting interval.

It is apparent from Equation (1) that, if the counting interval is constant, the average counting rate  $\overline{R}'$ , will always be a constant fraction of the initial counting rage,  $R'_0$ . Therefore, if the resolving time of the counter is neglected, the counting interval may be of any constant duration whatever without affecting the ratio of successive counts or the calculated value of the half-life.

However, since all counters have resolving times, it is necessary to evaluate the difference between the "true" average counting rate,  $\overline{R}$ , and the corrected average counting rate,  $\overline{R}$ , and the resolving time,  $\tau$ , as follows:

$$R = \frac{R''}{1+R'' \tau}$$
(2a)

$$R'' \frac{R}{1 - R \tau}$$
(2b)

$$\tau = \frac{1}{R} - \frac{1}{R''}$$
(2c)

The average observed counting rate,  $\overline{R}$ , is related to the instantaneous recorded counting rate, R, as follows:

Total observed count = 
$$\overline{R}t$$
 =  $\int_{0}^{t} Rdt = \int_{0}^{t} \frac{R''dt}{1 + R'' r}$   
=  $R_{0}'' \int_{0}^{t} \frac{e^{-\lambda t} dt}{1 + R_{0}'' r e^{-\lambda t}}$   
 $\overline{R} = 1 \log_{e} 1 + R'' 0 r$ 

 $1 + R_0^{"} \tau e - \lambda t$ 

λrt

(3)

R

 $\overline{R}$ " can now be evaluated from equations 2b and 3. Comparison of  $\overline{R}$ ! and  $\overline{R}$ " for various selected values of the constants reveals that there is a small but significant difference between the "true" counting rate and the corrected observed counting rate. The difference increases exponentially with an increase in the counting interval as well as with an increase in the average observed counting rate,  $\overline{R}$ . Such a relationship suggests that the deviation may be treated as an apparent increase in the value of the resolving time,  $\tau$ .

The apparent increase in the value of r may be conveniently determined from Equation (2c). The difference between the reciprocals of  $\overline{R}$  and  $\overline{R'}$  is then r':

$$r' = \frac{\lambda \tau t}{\log_e \frac{1 + R_0'' \tau}{1 + R_0'' \tau e^{-\lambda t}}} - \frac{\lambda t}{R_0' (1 - e^{-\lambda t})}$$
(4)

Evaluation of Equation (4) for selected values of the constants shows that r' is a constant for any given counting interval. Furthermore, the difference between r and r' increases as the square of the counting interval. This relationship would permit the introduction of a correction factor for data obtained with a counting interval which is a large fraction of the half-life. Such a correction is not necessary, however, if the variational method for computing the half-life is used.

In the variational method,  $\tau$  is treated as one of the unknown constants in a least-squares determination of the half-life. The slope and intercept of the semi-logarithmic decay curve are computed for each of several values of  $\tau$ , and the set of constants producing the best fit (lowest probable error) is selected as containing the most probable value of the half-life. Although the resolving time corresponding to this halflife is actually  $\tau'$ , rather than  $\tau$ , the difference is negligible for ordinary counting purposes.

Table 4 shows the results of evaluating Equations (1) through (4) for selected values of  $R'_0$  and t. The decay constant,  $\lambda$ , was selected to correspond approximately to the decay constant of lead-211; the resolving time,  $\tau$ , corresponds to that of the Nuclear Measurements Corp. PC-1 alpha counter. It will be noted that, even in the extreme case where the counting interval is equal to the half-life, the apparent increase in  $\tau$  is only four per cent. Furthermore, at the upper level of the practical counting range,  $(R'_0 = 2 \times 10^6 \text{ counts per minute})$ , the relative counting error is only 0.5 per cent.

1

The results in Table 4 indicate that the computed value of the half-life of lead-211 will be unaffected by the length of the counting interval used, so long as that interval is constant throughout the run. To test the validity of the assumptions upon which the derived equations are based, a series of runs is being made to obtain data under actual operating conditions.

Five runs have been made on the NMC PC-1 counter with one-minute counting intervals, one with a 5.12minute counting interval, and one with a 30-minute counting interval. A single run, with a five-minute counting interval, has been made on the NMC PC-3A counter. The data have been turned over to the Computer Analyses Group for least squares determination of the half-life and resolution times.

## Table 4

| t = 2 minutes       |           |           |           | :                       |                              |
|---------------------|-----------|-----------|-----------|-------------------------|------------------------------|
| R' or R'            | R.        | Ŕ         | R"        | <b>R</b> ' + <b>R</b> " | τ' <i>τ</i>                  |
| (cts/min)           | (cts/min) | (cts/min) | (cts/min) | (cts/min)               | (min)                        |
| 2 × 10 <sup>6</sup> | 1,962,085 | 1,640,225 | 1,962,044 | 41                      | 0.00011<br>× 10*7            |
| 1 × 10°             | 981,043   | 893,386   | 981,030   | 13                      | 0.00013                      |
| 5 × 10 <sup>5</sup> | 490,521   | 467,582   | 490,518   | 3                       | 0.00013                      |
| 2 x 10 <sup>5</sup> | 196,209   | 192,432   | 196,208   | . 1                     | 0.00014                      |
| t = 10 minutes      | 5         |           |           | •                       |                              |
| 2 x 10 <sup>6</sup> | 1,819,713 | 1,538,942 | 1,818,853 | 860                     | 0.0026<br>× 10 <sup>-7</sup> |
| 1 x 106             | 909,857   | 833,781   | · yu9,623 | 234                     | 0.0028                       |
| 5 × 10 <sup>s</sup> | 454,928   | 435,077   | 454,867   | 61                      | 0.0029                       |
| 2 × 10 <sup>5</sup> | 18 1,97 1 | 178,710   | 181,962   | 9                       | 0.0028                       |
| 1 × 10 <sup>5</sup> | 90,986    | 90,163    | 90,983    | 3                       | 0.0031                       |
| t = 36.1 minut      | es        |           |           |                         |                              |
| 2 × 10 <sup>6</sup> | 1,442,695 | 1,255,309 | 1,435,510 | 7 185                   | 0.035<br>× 10-7              |
| 1 × 10 <sup>6</sup> | 721,348   | 671,142   | 719,426   | 1922                    | 0.037                        |
| 5 × 10 <sup>3</sup> | 360,674   | 347,654   | 360,176   | 498                     | 0.038                        |
| 2 × 10 <sup>5</sup> | 144,270   | 142, 139  | 144,188   | 82                      | 0.039                        |
| 1 × 10 <sup>5</sup> | 72,135    | 71,598    | 72,114    | 21                      | 0.039                        |
| 5 x 10 <sup>4</sup> | 36,067    | 35,934    | 36,063    | 4                       | 0.031                        |

# COMPARISON OF TRUE AND OBSERVED COUNTING RATES FOR VARIOUS COUNTING INTERVALS<sup>a</sup>

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a - Half-life = 36.1 min; resolving time =  $1 \times 10^{-7} \text{ min}$ .

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** Additional determinations have been made on the density of liquid cerium metal by the vacuum pycnometer method using tantalum pycnometers of about  $0.5 \text{ cm}^3$  capacity. The density values at  $825^{\circ}$ C may be low since it was discovered that the pycnometers were not being lowered far enough into the melt to completely fill. After modification of the technique, cerium metal extruded from the pycnometer used at  $850^{\circ}$ , which indicated that the pycnometer was filled and, also, that the cerium must expand when solidifying at the melting point. Further density determinations are planned for liquid cerium, lanthanum, and praseodymium.

| Temperature (°C) | Density (g/cm³ |
|------------------|----------------|
| 825              | 6.58           |
|                  | 6.64           |
|                  | 6.57           |
| 850              | 6.69           |
| 875              | 6.48           |
| 900              | 6.66           |

#### DENSITY OF MOLTEN CERIUM

The 155.938 grams of 99.95 per cent pure plutonium metal was sealed in a tantalum capsule and suspended in the viscometer. The plutonium employed is from the same lot as used in a viscosity determination made at Mound Laboratory in July, 1960. The viscosity was investigated at eight different temperatures over a range from 648 to  $950^{\circ}$ C. Initially, the container with the plutonium was kept for 12 hours at  $750^{\circ}$ C. Thereafter, the sample was equilibrated for five to six hours at each temperature between viscosity determinations. The pressure within the apparatus was maintained between 4.4 x  $10^{-6}$  mm mercury.

The composite data from the determination of July, 1960 and the present determination are given below. Figure 1 illustrates the viscosity of plutonium as a function of absolute temperature. The line drawn represents a least squares solution for all 14 points reported.

| Run<br>No. | Temperature<br>℃ | Viscosity<br>Centipoise |
|------------|------------------|-------------------------|
| 2          | 950              | 3.97                    |
| 1          | 917              | 4.03                    |
| 2          | 902              | 4.11                    |
| 2          | 859              | 4.27                    |
| 1          | 824              | 4.43                    |
| 2          | 809              | 4.51                    |
| 2          | 765              | 4.66                    |
| 1          | 739              | 4.90                    |
| 2          | 712              | 5.10                    |
| 1          | 699              | 5.55                    |
| 2          | 670              | 5.46                    |
| 1          | 650              | 6.00                    |
| 2          | 648              | 6.02                    |
| 1          | 648              | 5.92                    |

#### VISCOSITY OF MOLTEN PLUTONIUM

Run 1 - July, 1960 Run 2 - January, 1961

After completion of the plutonium viscosity determination, the tantalum capsule was cooled, in vacuo, from the last temperature investigated (648°C). When the capsule was removed from the viscometer, it was found to be ruptured as shown in Figure 2. Deformations have been noted in previous viscosity determinations of plutonium, but not to the extent shown here.

High purity graphite liners have been fabricated for 0.75 inch and 0.625 inch diameter tantalum crucibles. These liners will be evaluated with lead from its melting point at  $327^{\circ}$ C to  $1000^{\circ}$ C.

High Temperature Calorimeter The construction of the high-temperature calorimeter has continued. Although the resistance wires for the nickel block heaters had previously been wound on their ceramic forms, it was necessary to remove the wire since the grooves were not of sufficient depth. A carbide tool bit was used to cut the grooves to a depth of 0.020 inch which was adequate to permit the wire to lie well below the surface of the ceramic.

The vacuum system for the calorimeter has been assembled and leak tested. With the use of a mechanical fore-pump and an oil diffusion pump, a reduced pressure of  $1.5 \times 10^{-5}$  mm. mercury can be maintained within the apparatus.

The three heater units of the furnace for the calorimeter have been connected to their respective controllers and thermocouples. The thermocouples have been located within the furnace as close as possible to the heater units. A water-cooled heat reflector is being constructed to shield the alpha-box from the radiation of the furnace.







Metallography An alpha-box is being designed to completely enclose the vacuum hot stage and ion pump. A device has been constructed to permit time lapse photography with the 16 mm movie camera sighting through the hot-stage microscope. The time interval between individual exposures can be varied from nine seconds to two hours.

Exploratory work continued with the cathodic etcher. At argon pressures greater than 200 microns, a stable arc was obtained and satisfactory etching occurred for 1013 steel, 304 stainless steel, and a cerium-copper alloy. Other materials will be investigated before plutonium alloys are introduced into the system. The recent difficulties with arc stability results from the fact that the specimen to be etched is recessed below the cathode surface. A complete shielding of the cathode appears to be necessary for good arc stability. Such shield is being fabricated from "Teflon". The anode has been shielded but with little beneficial effect.

**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.

Ten weight per cent plutonium oxide was incorporated into an experimental glass containing silicon dioxide, aluminum oxide, calcium oxide, sodium oxide and potassium oxide. Fibers were made from these and leach tests were run in room temperature water, 0.1 normal hydrochloric acid, 0.5 normal nitric acid, and hot water. Analyses of the leach solutions indicated, after 1557 hours in room temperature water, a loss of 0.099 per cent of the plutonium oxide, in 0.1 normal hydrochloric acid at room temperature after 1534 hours a loss of 0.181 per cent of the plutonium oxide, after 784 hours at room temperature in 0.5 normal nitric acid a loss of 0.380 per cent of the plutonium oxide, and after 784 hours in hot water a loss of 1.30 per cent of the plutonium oxide from the glass fibers. The high rate of attack of the fibers by the hot water apparently is the result of increased dissolution of sodium oxide from the glass. Becoming basic in character, the hot leach solution then attacks the vitreous portion of the material releasing additional plutonium oxide.

A glass of the same parent composition as the above experimental glass, but containing fifteen weight per cent dissolved plutonium oxide, was previously homogenized and drawn into fibers. Leaching tests on these fibers in room temperature water are now in progress. The first results, after two days leaching, indicate a loss of plutonium oxide from the glass fibers comparable to that obtained for the ten weight per cent plutonium oxide composition after an equivalent period of leaching.

An experimental glass composition containing 56 weight per cent silicon dioxide, 24 weight per cent sodium oxide and 20 weight per cent plutonium oxide has been successfully homogenized and fabricated into fibers. The formulation was homogenized after extensive thermal treatment at approximately 1650°C. If higher temperatures could be employed, more plutonium oxide could undoubtedly be dissolved in the melt. However, twenty weight per cent appears to be near the limit of plutonium oxide which can be dissolved in this composition and successfully drawn into fibers without devitrification. The liquidus temperature of equilibrium crystallization of plutonium oxide for the composition appears to be a little under 1375°C which is the holding temperature for the molten glass during fiber drawing.