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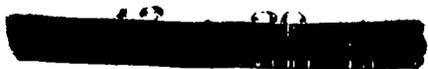
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ALLOYS OF PLUTONIUM WITH ALUMINUM

Work done by:

Written by:

- F. H. Ellinger
- M. Gibbs
- F. I. Newville
- R. D. Moeller
- F. W. Schonfeld
- J. Singer
- V. O. Struebing
- C. R. Tipton, Jr.
- P. Vigil
- D. D. Whyte

- R. D. Moeller
- F. W. Schonfeld

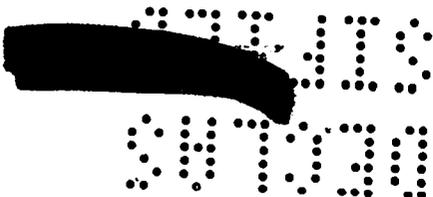
Edited by:

A. S. Coffinberry



PLUTONIUM TECHNOLOGY

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ABSTRACT

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A. On the basis of the incomplete data available to date, the following tentative suggestions are made concerning features of the equilibrium diagram of the plutonium-aluminum system:

1. Essentially zero solid solubility of plutonium in aluminum at all temperatures.
2. A eutectic composition of 98.3 atomic percent aluminum. The eutectic temperature is 647°C and the two phases involved are pure aluminum and an intermediate phase.
3. An intermediate phase of complex crystal structure in the composition region  $\text{PuAl}_4$ .
4. A second intermediate phase of complex crystal structure corresponding to  $\text{PuAl}_3$ , which may react peritectically to form the intermediate phase in the region of  $\text{PuAl}_4$ .
5. A third intermediate phase corresponding to the formula  $\text{PuAl}_2$ , which may react peritectically to form  $\text{PuAl}_3$ . The structure of  $\text{PuAl}_2$  has been established as cubic of the  $\text{Cu}_2\text{Mg}$  type, and is isomorphous with  $\text{UAl}_2$ , with  $a_0$  equal to 7.820 kX. Its melting point is thought to be the highest of the system and to lie between 1100°C. and 1300°C.

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6. A fourth intermediate phase which exists in the region of PuAl. It appears to have a structure distorted from cubic, possibly tetragonal with an axial ratio of about 0.98. This phase may result from a peritectoid reaction at about 585° between the intermediate phase PuAl<sub>2</sub> and the delta solid solution (solid solution of aluminum in delta plutonium, stable at room temperature).

7. A fifth intermediate phase, which may have a tetragonal crystal structure, corresponding to the formula Pu<sub>3</sub>Al, probably results from a peritectoid reaction at about 565°C between the intermediate phase PuAl and the delta solid solution.

8. The upper limit of solubility of aluminum in delta plutonium at temperatures between 25°C and 30°C is in the neighborhood of 12.5 atomic percent aluminum. This solubility is evident only in the face-centered cubic delta phase. Solubilities in the alpha, beta, and gamma phases are zero percent aluminum while the limit of solubility in epsilon plutonium is unknown.

9. A two-phase field of alpha plutonium plus delta solid solution at aluminum percentages below about 2 atomic percent at room temperature. This field changes to beta plus delta, and gamma plus delta solid solution at successively higher temperatures. When the alpha-to-beta and

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beta-to-gamma transformations are observed there is no noticeable change in the temperature of transition.

10. The aluminum-rich portion of the phase diagram of the plutonium-aluminum system is apparently similar to the high aluminum regions of the uranium-aluminum and the rare-earth-aluminum systems, but somewhat more complex.

B. Alloys containing more than 80 per cent of the face-centered cubic delta solid solution (2-20 atomic per cent aluminum) are workable both hot and cold, and possess good casting characteristics. The intermediate phase  $\text{Pu}_3\text{Al}$  is brittle at room temperature but exhibits moderate plasticity at temperatures above  $400^\circ\text{C}$ . The intermediate phase  $\text{PuAl}$  is brittle at room temperature but is quite plastic at temperatures above  $450^\circ\text{C}$ . The intermediate phases  $\text{PuAl}_2$ ,  $\text{PuAl}_3$ , and  $\text{PuAl}_4$  are brittle and remain brittle up to  $485^\circ\text{C}$ , the maximum temperature at which their forming characteristics have been investigated. Castabilities of alloys containing between 40 and 90 atomic percent aluminum were found to be poor for several reasons (explained below). Alloys containing from 90 to 100 atomic per cent aluminum are castable but are subject to a high degree of solidification shrinkage and are only moderately workable. These alloys possess excellent machinability, however.

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C. No systematic data on corrosion are yet available, but the resistances of the alloys to corrosion in laboratory atmosphere at room temperature appear, qualitatively, quite good. The alloys containing more than about 75 atomic percent aluminum seem to be very resistant to corrosion in laboratory atmosphere at room temperature.

D. Although results of experimental measurements are not yet available,\* a few deductions may be made regarding thermal conductivities to be expected for some of the plutonium-aluminum compositions. These point to conductivities of the order of that for pure aluminum in the 90 to 100 atomic percent aluminum range, about one-third that of stainless steel in the 2 to 20 atomic percent aluminum range, and very poor thermal conductivities (characteristic of intermetallic compounds) for compositions between about 25 to 85 atomic percent aluminum.

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\* Since this report was written, preliminary measurements of thermal conductivities have indicated values from about one-half to equal that of pure aluminum for the composition range 90 to 100 atomic percent aluminum.

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INTRODUCTION

On 1 July 1949 Group CMR-5 was authorized to undertake an investigation of the plutonium-aluminum system. Since that time work has progressed concurrently with other researches. A recent intensification of interest has made advisable an accelerated work program and an immediate presentation of such data as are now available. One should keep in mind that suggestions and conclusions presented are tentative, unless otherwise specified, and may be modified at a later date.

Alloy compositions are expressed as atomic per cent. Where results of chemical analysis are available, these results are also shown. In most of the following discussion, however, alloy compositions are designated by the nominal atomic percent aluminum content aimed for in the original preparation of the alloy.

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## GENERAL EXPERIMENTAL PROCEDURES

The various methods of attack that have been utilized involved the standard techniques of physical metallurgy with such modifications and additions as were made necessary by the reactivity and toxicity of plutonium.

The alloys were prepared by vacuum-melting. The buttons obtained through the melting operation were sampled for chemical analyses and then divided into two specimens. One specimen, in the as-cast condition, was utilized in microstructural investigations which consisted of visual examination, photomicrography, micro-hardness, and micro-lineal analyses to determine percentages of phases present. The second half of the button was cold-worked, when possible, and equilibration heat-treated. This specimen was utilized for structural investigations by means of x-ray diffraction. The remaining portion of each heat-treated specimen was later examined microscopically, so that structures in both the as-cast and heat-treated conditions might be compared.

In the determination of liquidus and solidus temperatures, both inverse-rate and time-temperature curves were obtained, the former manually and the latter autographically.

In order to obtain estimates of the workabilities of the various alloys, some were cold-forged, some were cold-

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rolled, several were hot-pressed, and three aluminum-rich alloys were hot-extruded. The extruded rods were prepared for use as specimens in the measurement of thermal conductivities. These measurements are being made by R. B. Gibney of Group CMR-9, and will be reported elsewhere.

While no systematic program for the determination of corrosion rates of plutonium-aluminum alloys has been undertaken, the oxidation behavior of alloy ingots at room temperature in laboratory atmosphere has been noted, thus enabling some conclusions to be drawn regarding their general corrosion resistance in ordinary environments.

## MELTING AND CASTING

The alloying and casting of specimens for all solid-state investigations were accomplished by vacuum-melting. Vacua of the order of  $10^{-4}$  mm of Hg were maintained throughout all melting operations. Accurately weighed amounts of the two components were placed in the melting crucible inside a resistance furnace which was surrounded by a brass "vac-can." The crucibles were magnesium oxide compacted with magnesium sulfate binder and fired at  $1950^{\circ}\text{C}$ . Before use, the crucibles were degassed at  $1100$  to  $1200^{\circ}\text{C}$ . Metal weights were so chosen as to yield buttons of about  $0.3$  cubic centimeter in size, small enough to avoid gross segregation and at the same time large enough so that the composition might be weighed out with a reasonable degree of accuracy.

The melting stocks utilized were high-purity aluminum, obtained from the Aluminum Company of America, and remelted plutonium stock RZ-16. For analyses of these materials, see Appendix I.

The melting cycle first utilized was to heat rapidly to about  $1125^{\circ}\text{C}$ , hold for five minutes, and cool at the natural rate of the furnace (approximately  $5^{\circ}/\text{min.}$ ). This procedure was found to be satisfactory for production of all

alloys except those containing from 50 to about 80 atomic per cent aluminum. Such alloys gave evidence of incomplete melting at temperatures up to 1250°C. When higher temperatures were employed (around 1300°C) the alloys tended to form brittle, porous "clinkers" with evidence of excessive spattering. Perfect alloying has not yet been attained within this composition range. It is believed that the relatively high vapor pressure of aluminum (70 microns at 1125°C) may result in undesirable shifts of composition at high temperature and thus further complicate alloying.

The castabilities of alloys containing from 90 to 100 atomic per cent of aluminum have been observed while producing extrusion slugs 1/2 inch in diameter by 2 inches in length. The eutectic composition had, of course, the best casting characteristics of the aluminum-rich alloys, but even in this case about 5 per cent solidification shrinkage occurred. The alloys seemed to possess good fluidity, but because of the presence of essentially pure aluminum, carefully controlled directional solidification was necessary to eliminate piping and other shrinkage cavities. The fluidity of the delta-phase solid solution was apparently high and its solidification shrinkage seemed small, although no large castings have been produced on which more adequate observations could be made. Alloys containing between 40

and 90 atomic per cent aluminum were found difficult to cast, primarily because of the high melting temperature of the compound  $\text{PuAl}_2$ . High shrinkage and brittleness at lower temperatures contributed to casting difficulties.

The densities of the alloys so far produced are listed in Table I and presented graphically in Figure 1. The solid line shown in Figure 1 is a curve of densities calculated by the rule of mixtures using 2.70 g/cc for pure aluminum and 15.85 g/cc, the density of the face centered cubic delta phase, for plutonium.

TABLE I

| Specimen Number | Weighed Out Composition (At. % Al) | Composition by Chemical Analysis (At. % Al) | Density As Cast (g/cc) |
|-----------------|------------------------------------|---|------------------------|
| 321             | 99.05                              | 98.89                                       | 2.93                   |
| 292             | 98.00                              | 98.07                                       | 3.08                   |
| 293             | 95.00                              | 95.06                                       | 3.60                   |
| 291             | 90.00                              | 90.23                                       | 4.43                   |
| 294             | 85.05                              | 85.62                                       | 5.18                   |
| 340*            | 82.50                              | 82.50                                       | 5.89                   |
| 312             | 80.01                              | 82.37                                       | 5.77                   |
| 341*            | 79.14                              |   | 6.05                   |
| 313             | 75.00                              | 77.05                                       | 6.18                   |
| 342             | 72.49                              |   | 6.78                   |
| 314             | 70.01                              | 69.20                                       | 7.39                   |
| 298             | 65.02                              |   | 7.61                   |
| 299             | 60.05                              | 58.90                                       | 7.16(?)                |
| 300             | 54.98                              |   | 9.92                   |
| 301             | 50.01                              |   | 10.08                  |
| 302             | 45.12                              |   | 10.84                  |
| 303             | 40.09                              |   | 11.13                  |
| 304             | 34.96                              |   | 11.90                  |

\*Composition doubtful, spattering occurred during melting.

TABLE I (Continued)

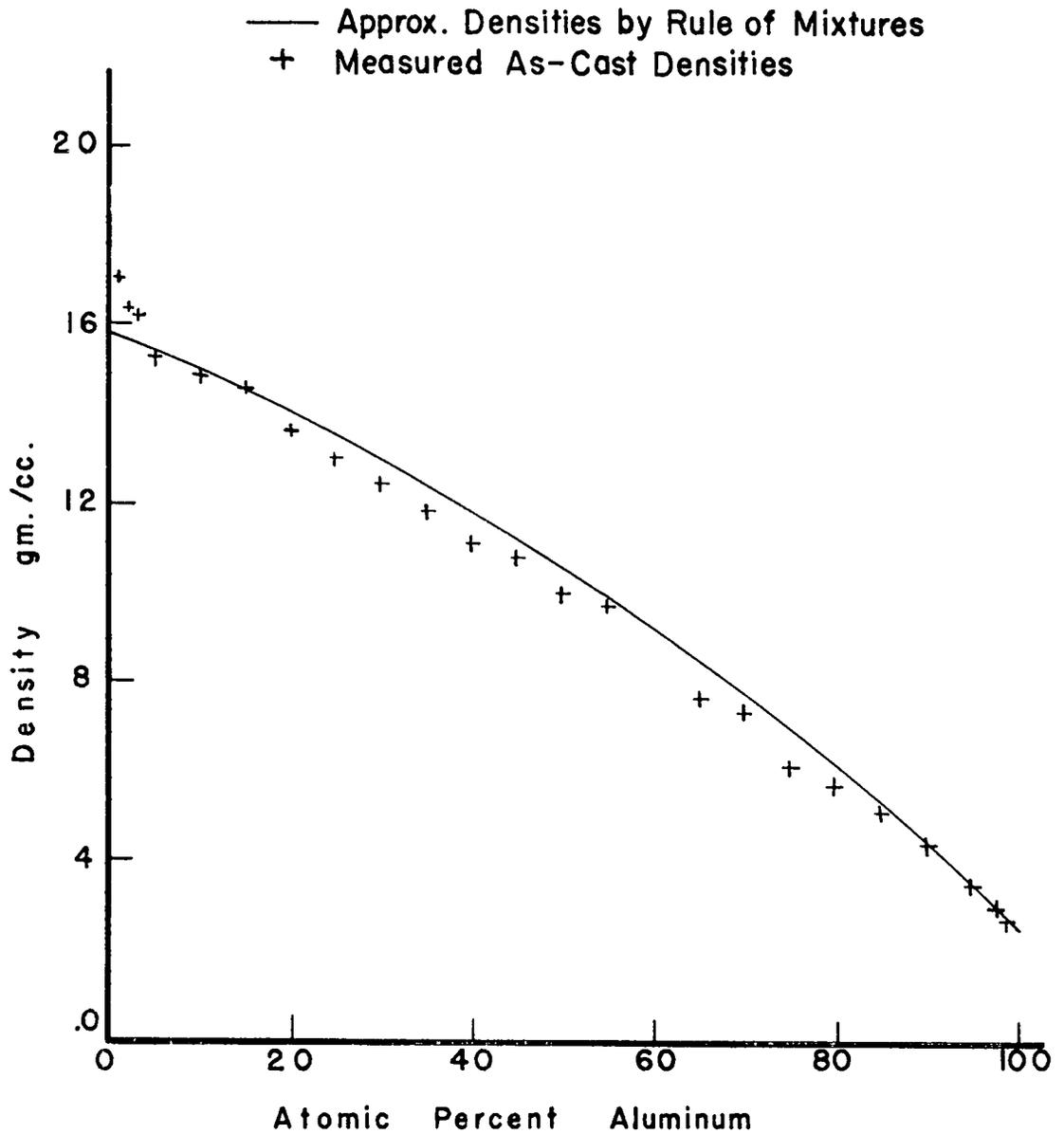
| Specimen Number | Weighed Out Composition (At. % Al) | Composition by Chemical Analysis (At. % Al) | Density As Cast (g/cc) |
|-----------------|------------------------------------|---|------------------------|
| 305             | 29.94                              |   | 12.53                  |
| 306             | 25.03                              |   | 13.02                  |
| 307             | 19.95                              |   | 13.71                  |
| 336             | 17.60                              |   | 14.31                  |
| 337             | 14.99                              |   | 14.60<br>(and 14.58)   |
| 308             | 15.00                              |   | 14.56                  |
| 338             | 12.51                              |   | 14.84                  |
| 309             | 10.08                              |   | 14.90                  |
| 315             | 4.99                               |   | 15.40                  |
| 316             | 2.92                               |   | 16.12                  |
| 317             | 1.99                               | 4.35**                                      | 16.36                  |
| 318             | 1.10                               | 2.94**                                      | 17.05                  |
| 296***          | 74.56                              | 67.23                                       | 8.36                   |
| 297***          | 70.04                              | 66.00                                       | 8.36                   |

\*\* Since chemical analyses were performed by determining weight per cent plutonium and obtaining weight per cent aluminum by difference, and since conversion from weight per cent aluminum to atomic per cent aluminum (for lower percentages ) multiplies errors by a factor of approximately 9, good agreement between weighed-out and analyzed compositions cannot be expected for low-percentage aluminum.

\*\*\* These were clinkers, taken to 1325°C in melting. Considerable weight loss occurred during melting.

Figure 1

VARIATION OF DENSITY WITH COMPOSITION  
OF PLUTONIUM - ALUMINUM ALLOYS



## THERMAL ANALYSIS

In order to establish the liquidus and solidus temperatures of the plutonium-aluminum system, a vacuum-melting furnace was employed for thermal analysis. By means of chromel-alumel thermocouples, time-temperature curves were obtained autographically on a Leeds and Northrup Model-S Micromax Recording Potentiometer, and inverse-rate curves were obtained manually through use of a Leeds and Northrup Type-K precision potentiometer. Both types of data were recorded simultaneously and during both heating and cooling portions of the thermal cycles. The specimens were prepared by additions of plutonium metal to an initial charge of 27 grams of aluminum.

Since, until very recently, interest in such data was not so high as in other features of the diagram, this work was not begun until 10 October 1949. Consequently, only four alloys (and pure aluminum) have so far been run. Data are presented graphically in Figures 2 through 8. Heating curves for the 90 and 95 atomic percent aluminum alloys are not as yet available. The curves obtained from the 99 atomic percent aluminum alloy are typical of results obtained from an alloy consisting of a primary phase plus a eutectic mixture. The discrete steps shown on the plateaus

of the time-temperature curves of the 98, 95, and 90 atomic per cent aluminum alloys are not explained, but may have resulted from the presence of an extremely narrow solid-plus-liquid field with an upper limit defined by a peritectic horizontal.

Several extremely small heat effects were noted at higher temperatures during runs on the 95 and 90 atomic per cent aluminum alloys, which points may have resulted from the presence of a second peritectic horizontal at about 725° C. The lower portions of the time-temperature curves obtained from these two alloys suggest a solid-state reaction extending over a range of temperature, and may represent the presence of a solvus line. If this is so, then the intermediate phase occurring at about 80 atomic per cent aluminum has a range of homogeneity which is rapidly narrowed with decreasing temperature.

The above remarks concerning the two peritectic horizontals and a solvus line must as yet be regarded as largely speculative.

Points well defined by thermal arrests are as follows:

| Weighed-Out<br>Composition<br>(At. % Al) | Liquidus<br>Temperature<br>(°C) | Solidus<br>Temperature<br>(°C) |
|--|---------------------------------|--------------------------------|
| 100                                      | 660.2                           | 660.2                          |
| 99.01                                    | 653                             | 647                            |
| 98.06                                    | -                               | 647                            |
| 95.02                                    | -                               | 647                            |
| 89.63                                    | -                               | 647                            |

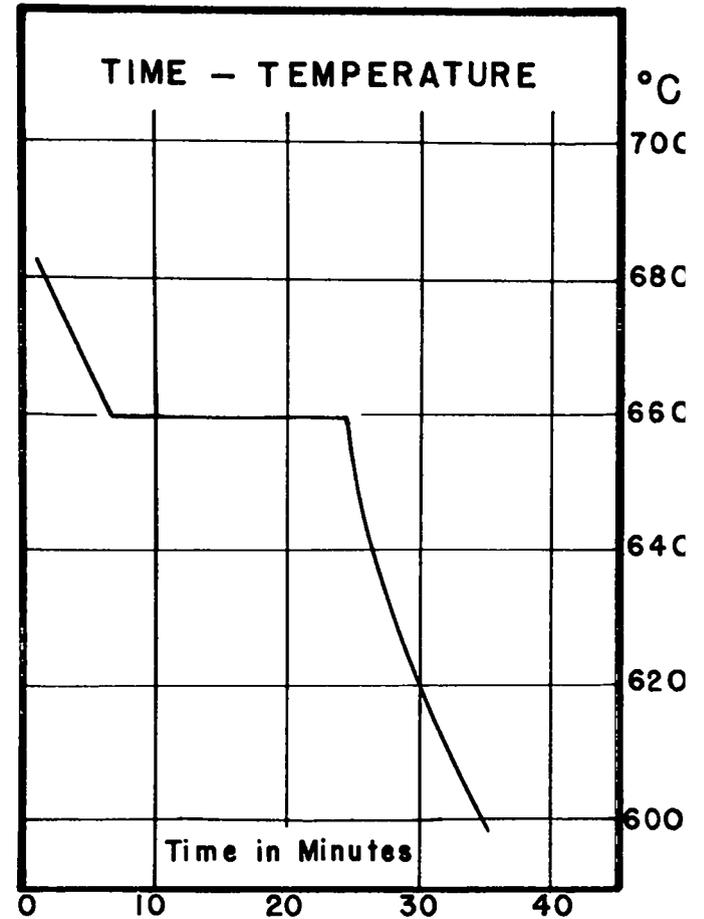
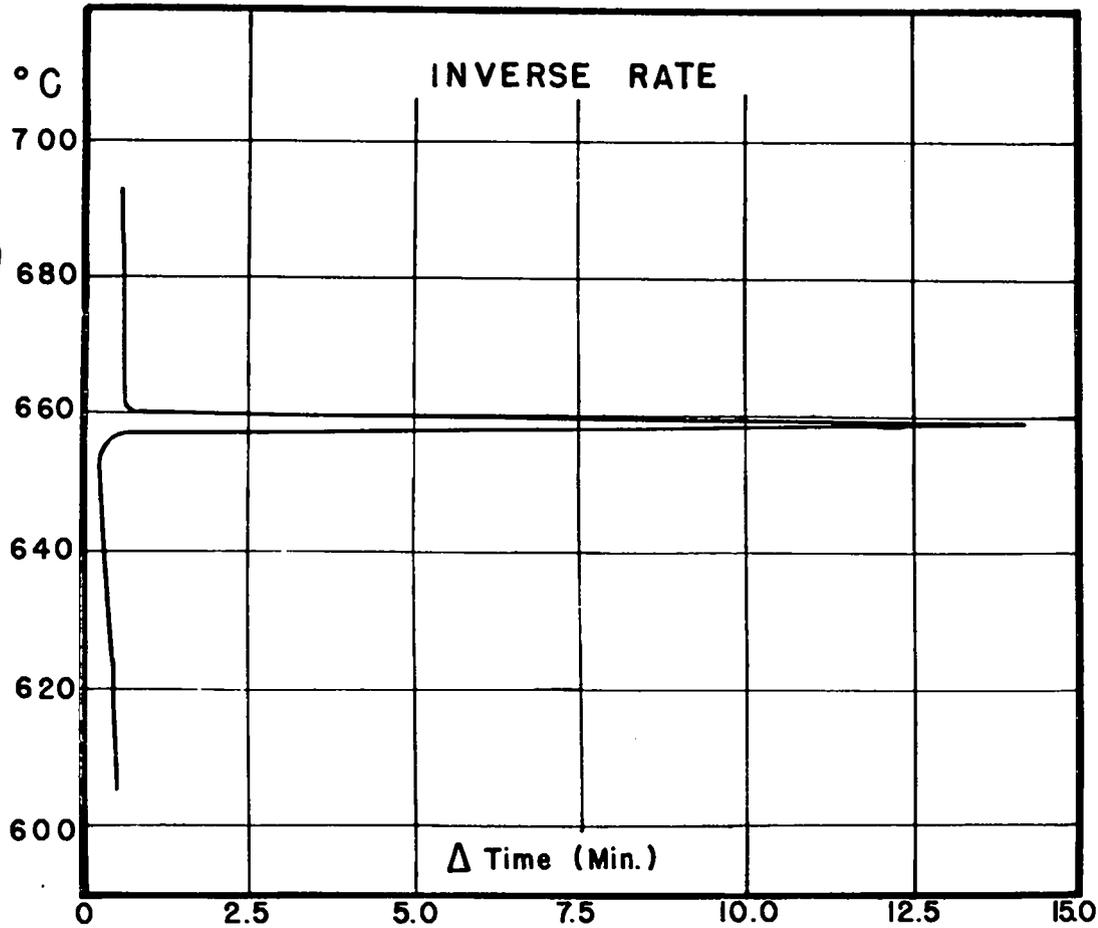
A plot of these data has set the eutectic composition at 98.3 atomic percent aluminum and the eutectic temperature at 647°C.

Points ill defined by thermal arrests (to be regarded as largely speculative) are as follows:

| Weighed-Out<br>Composition<br>(At. % Al) | PuAl <sub>4</sub><br>Solvus<br>Temp (°C) | PuAl <sub>4</sub><br>Peritectic<br>Temp (°C) | PuAl <sub>3</sub><br>Peritectic<br>Temp (°C) | Liquidus<br>Temp<br>°C |
|--|--|--|--|------------------------|
| 98.06                                    | 647 - 636                                | 652  | ---  | ---                    |
| 95.02                                    | 647 - 636                                | 652  | 725  | 820                    |
| 89.63                                    | 647 - 636                                | 652  | 725  | ---                    |

Figure 2

THERMAL ANALYSIS OF 100 % ALUMINUM  
COOLING CURVES

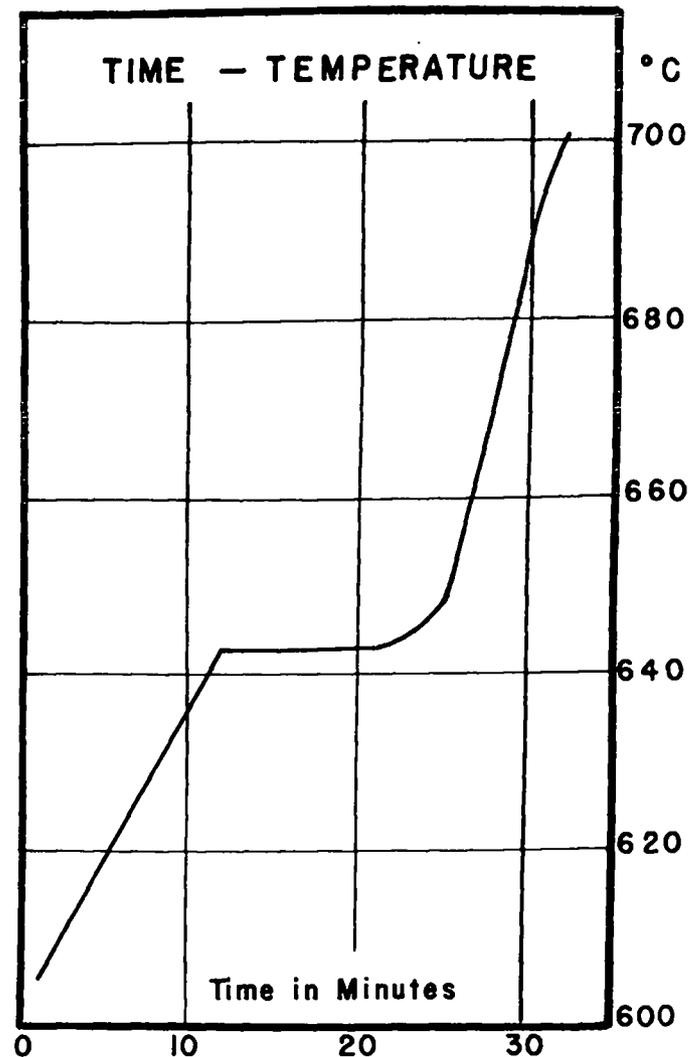
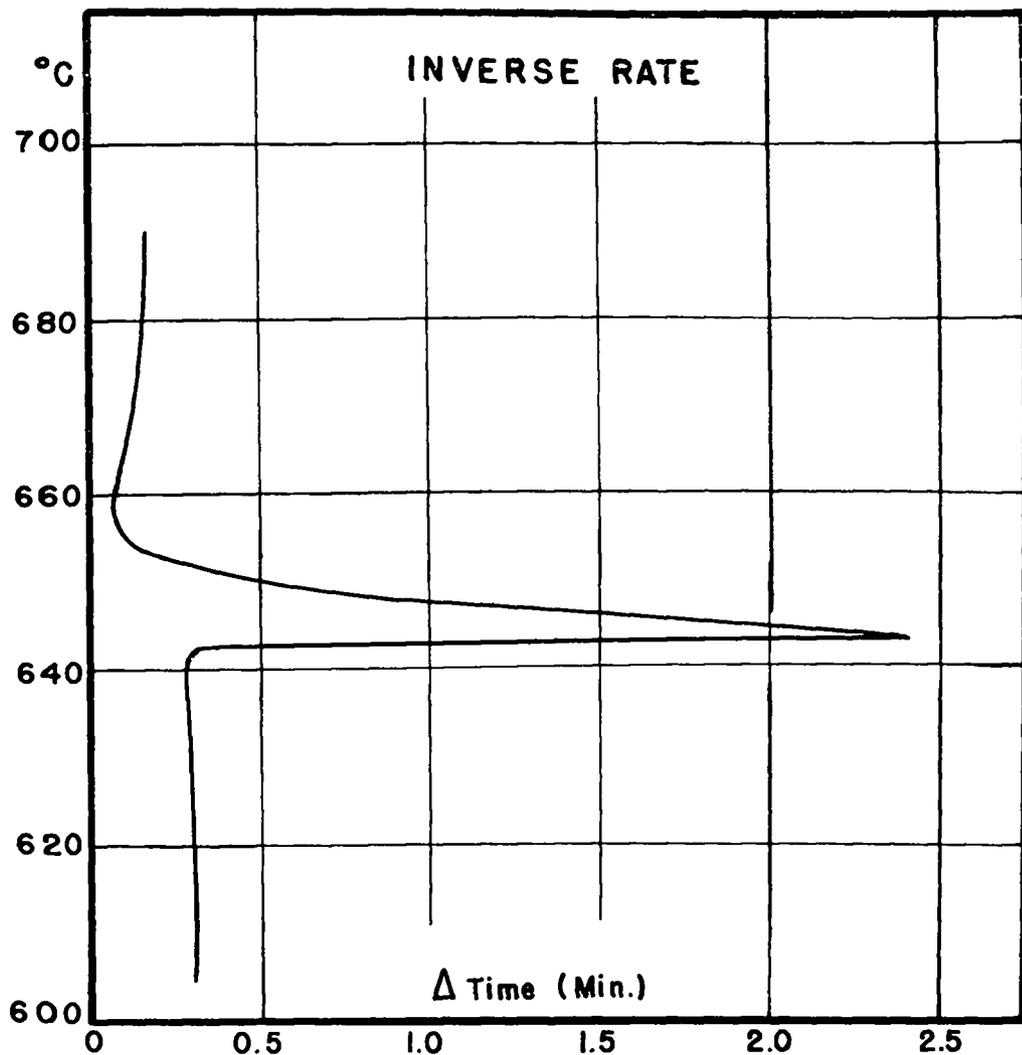


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Figure 3  
THERMAL ANALYSIS OF 99% AL, 1% PU ALLOY  
HEATING CURVES



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Figure 4  
 THERMAL ANALYSIS OF 99% AL, 1% PU ALLOY  
 COOLING CURVES

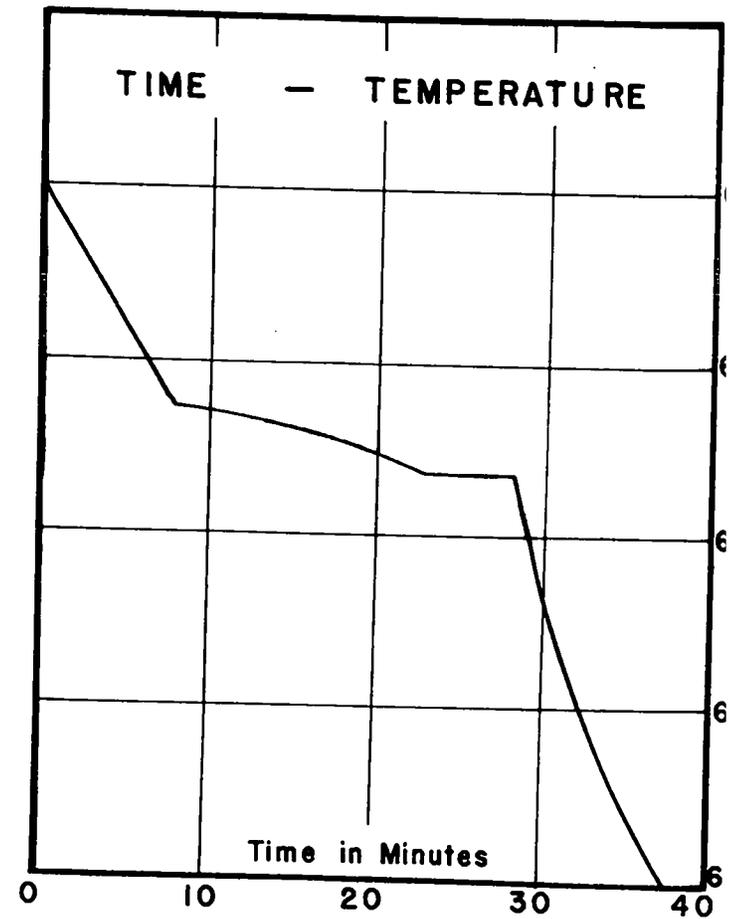
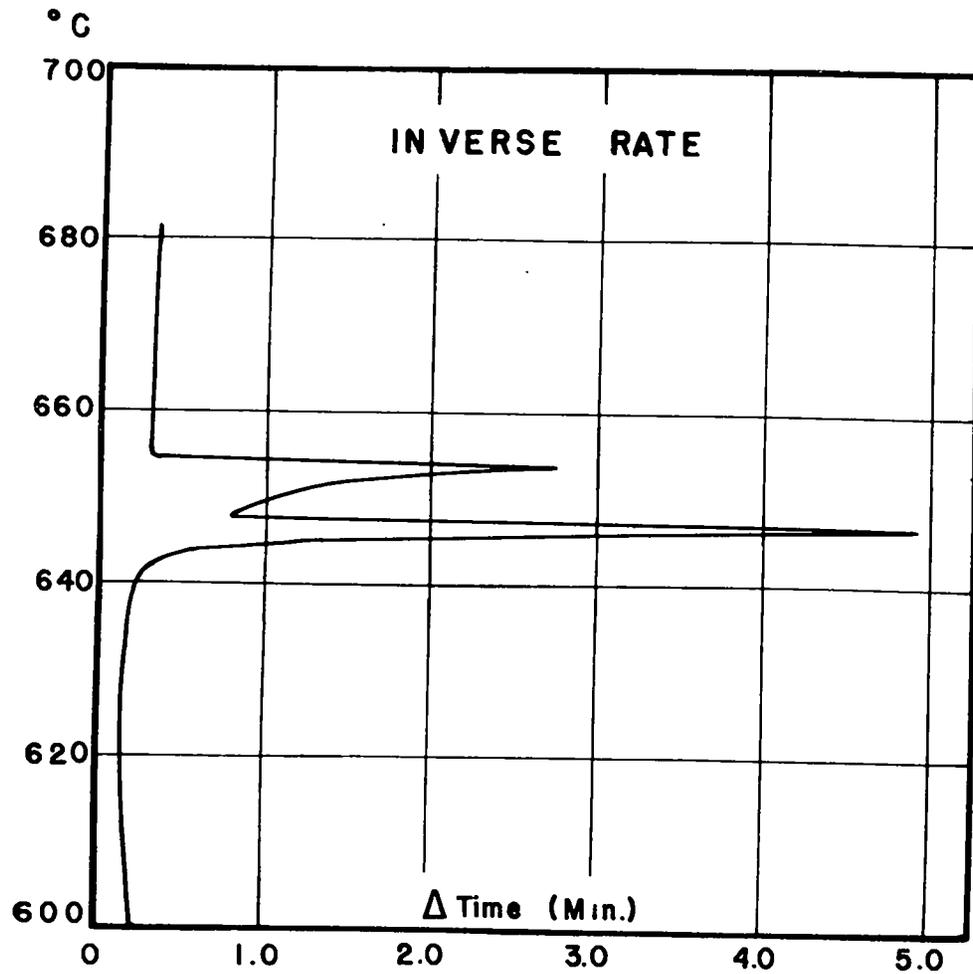
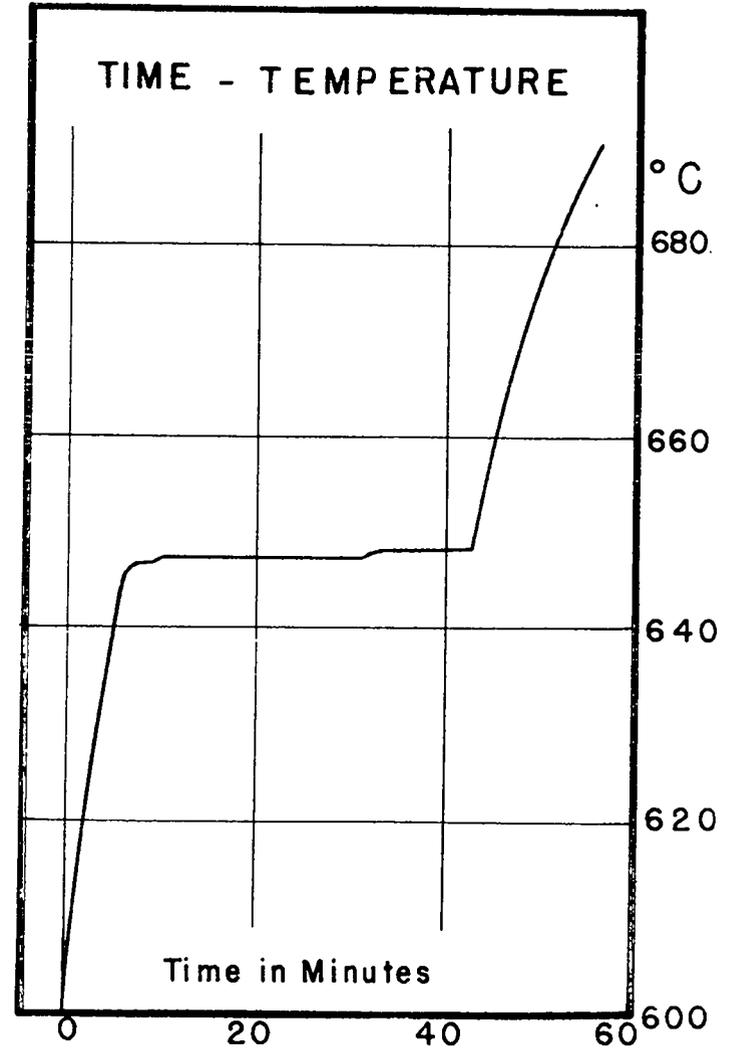
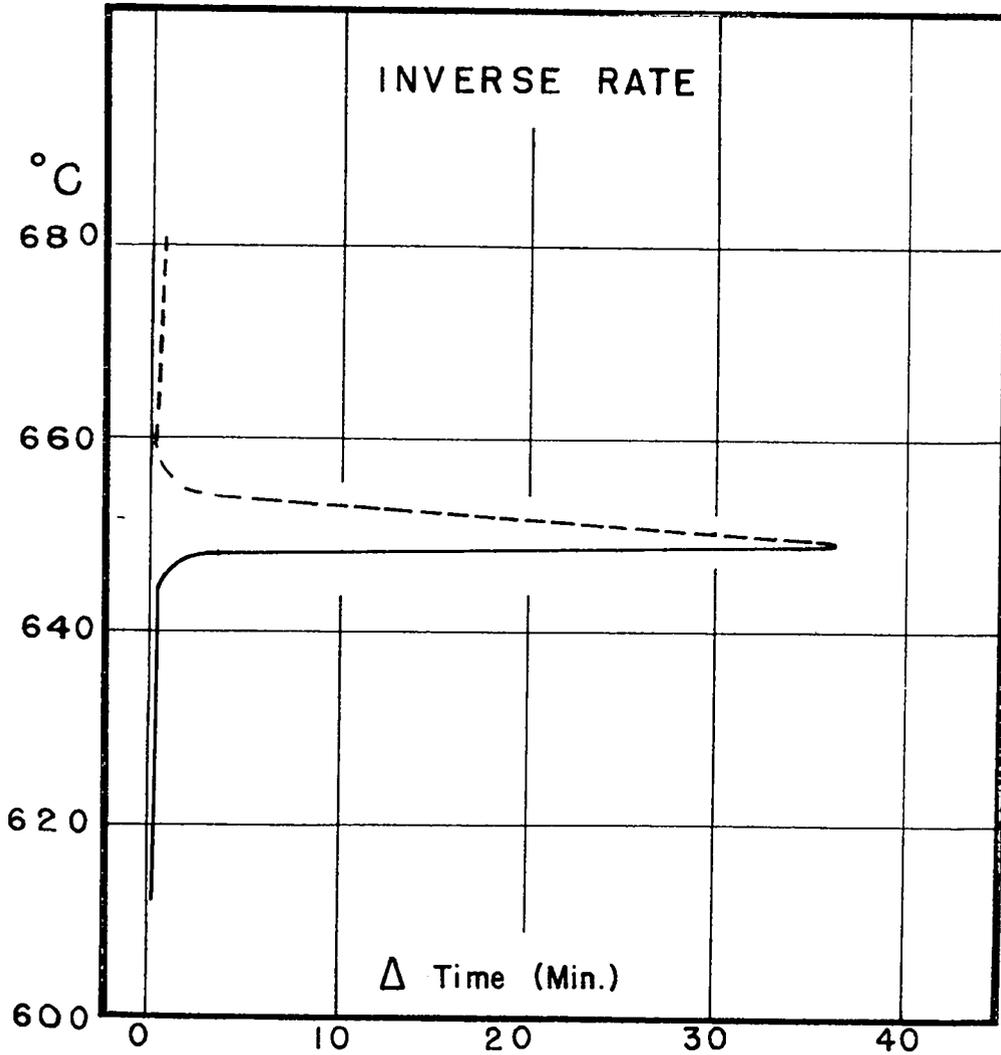


Figure 5

# THERMAL ANALYSIS OF 98% AL, 2% PU ALLOY

## HEATING CURVES



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Figure 6  
 THERMAL ANALYSIS OF 98% AL, 2% PU ALLOY  
 COOLING CURVES

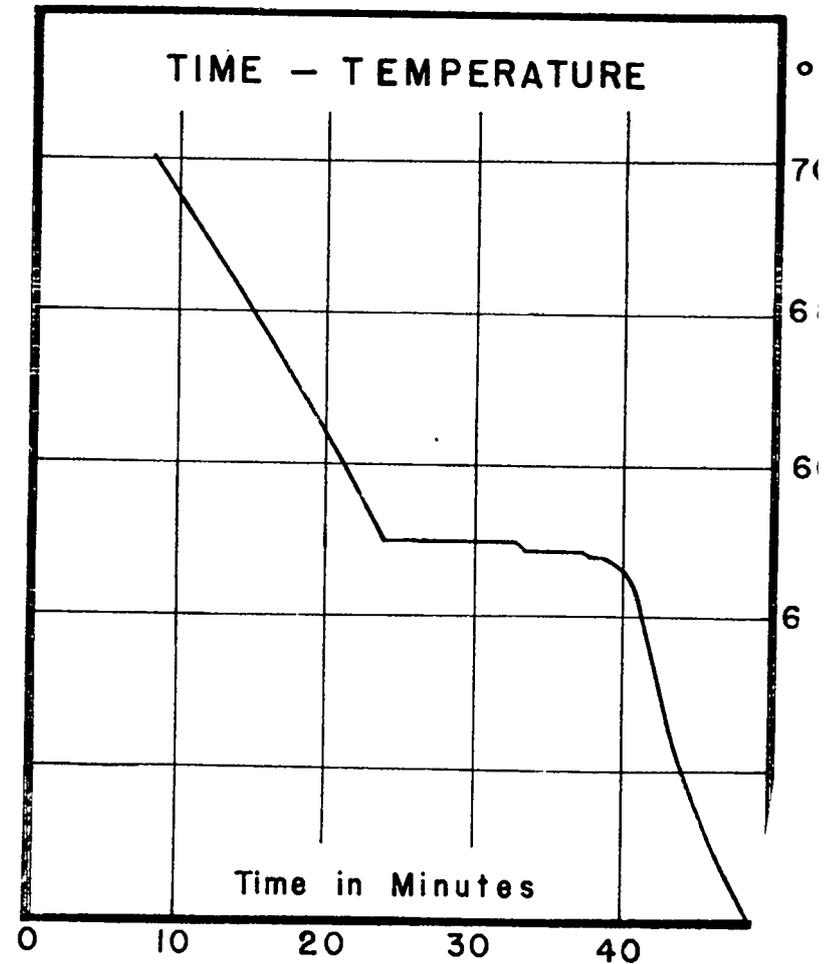
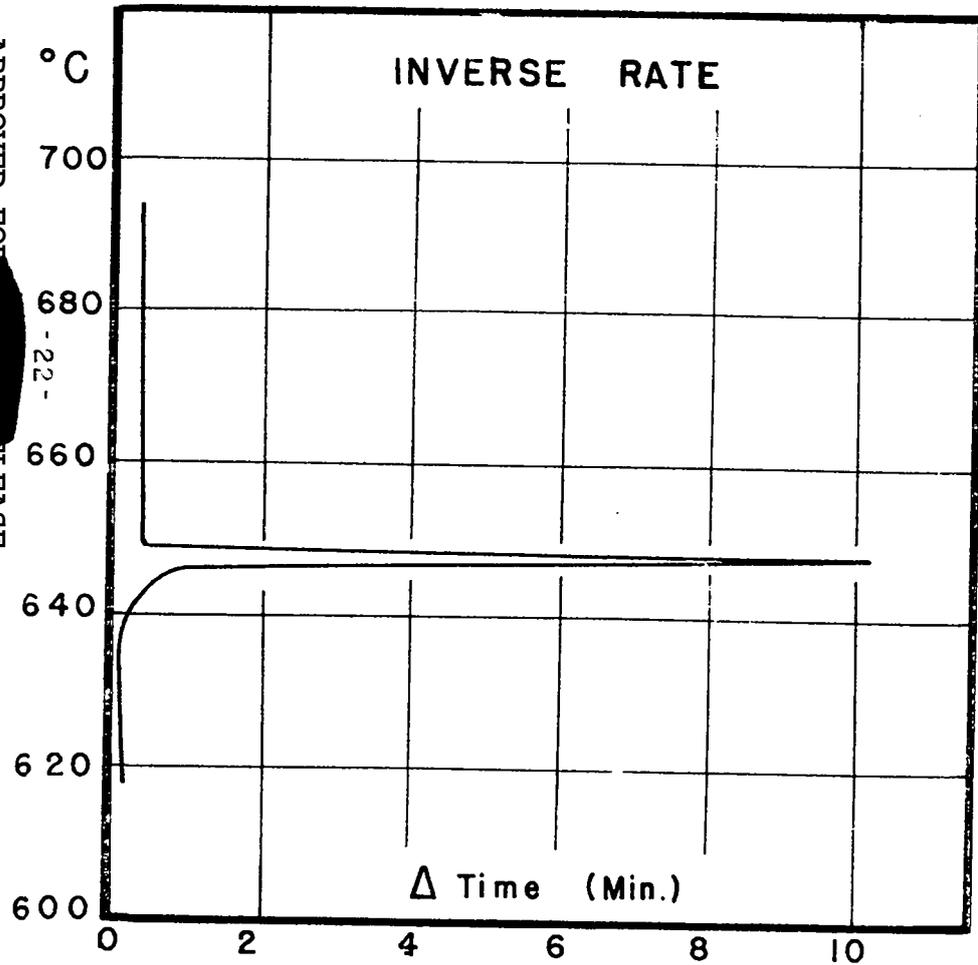
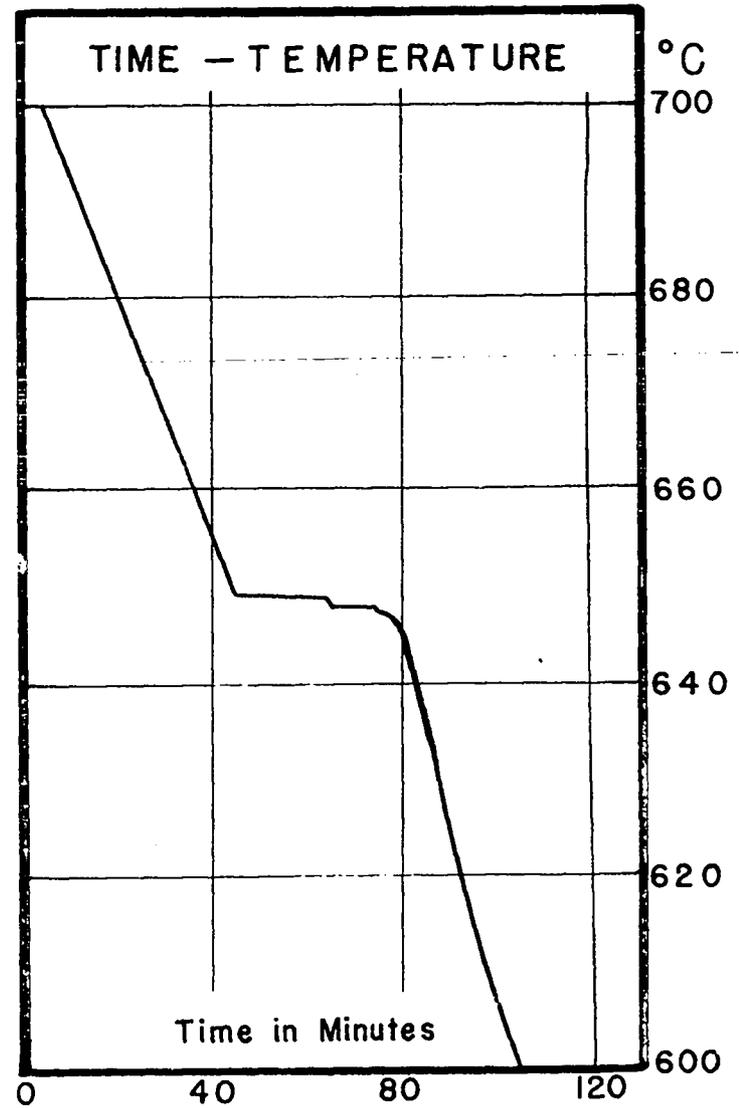
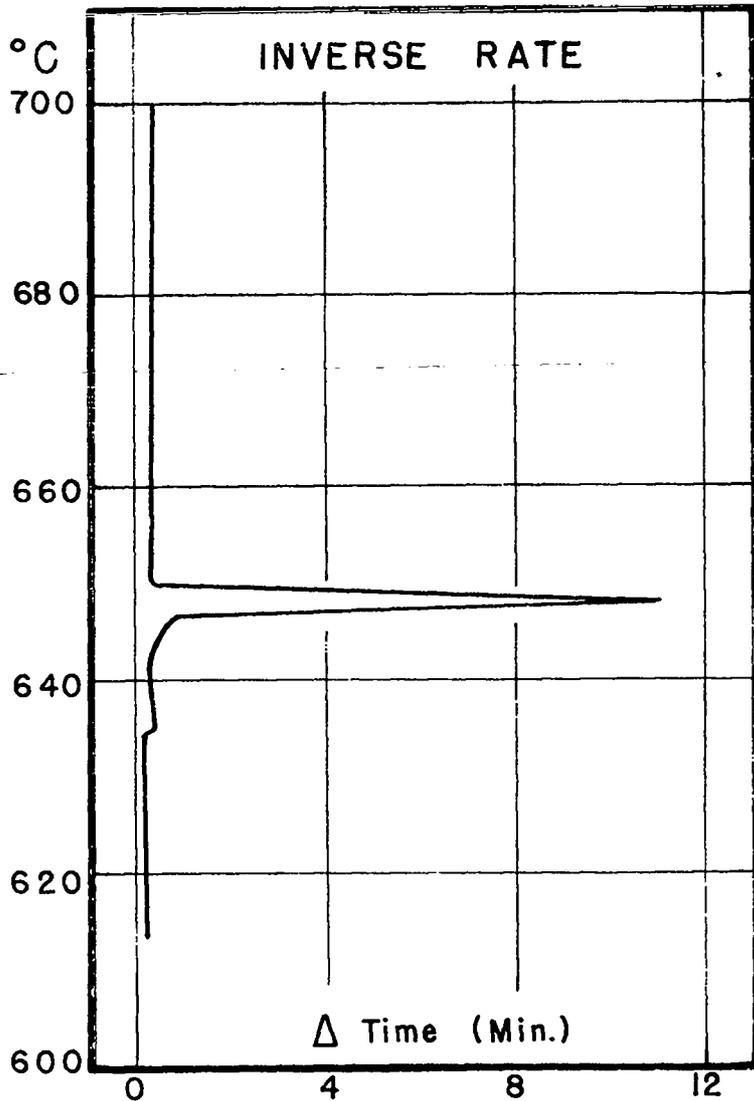


Figure 7  
 THERMAL ANALYSIS OF 95% AL, 5% PU ALLOY  
 COOLING CURVES



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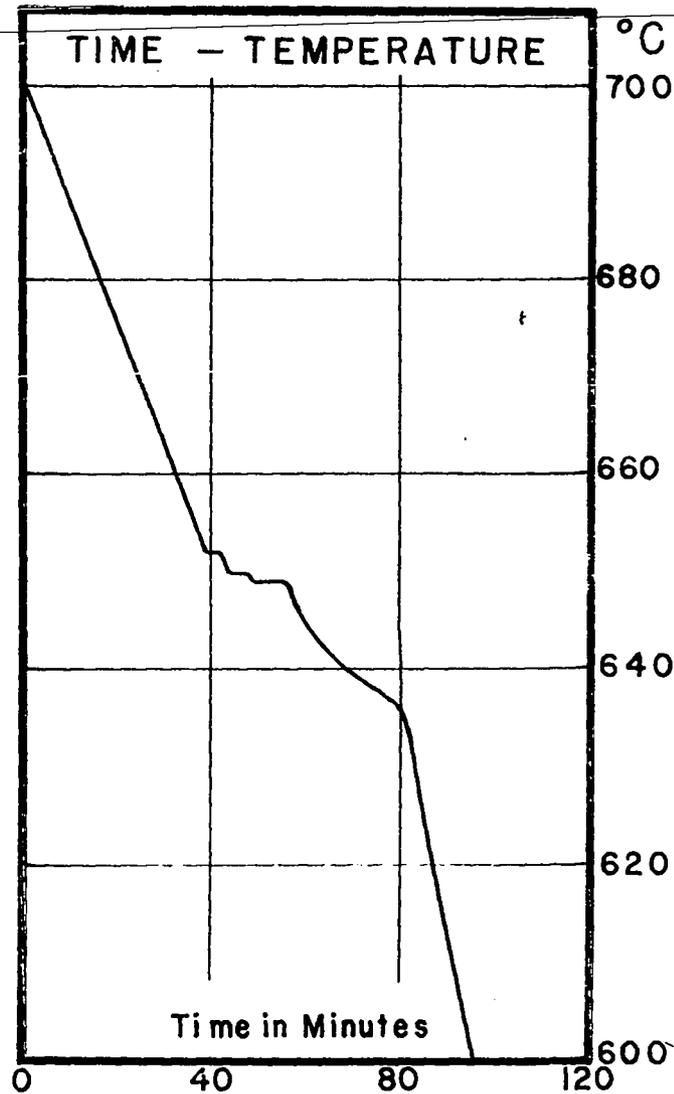
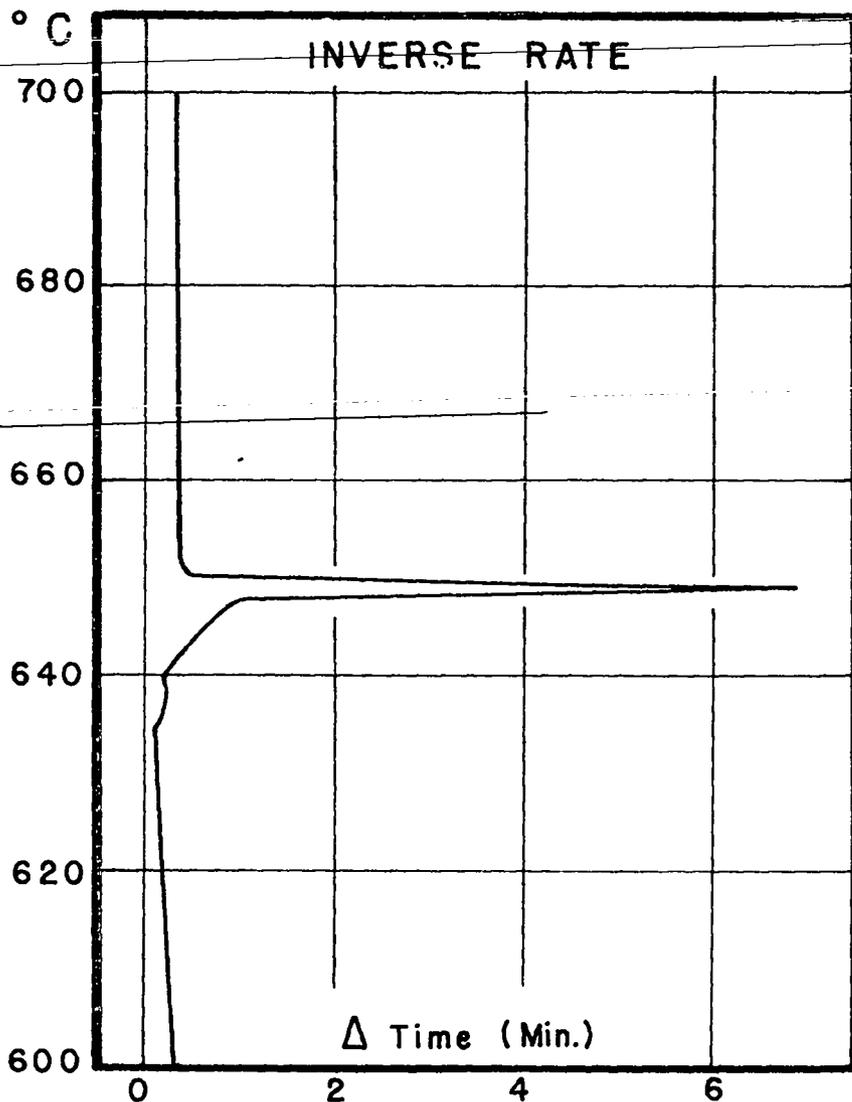
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Figure 8

# THERMAL ANALYSIS OF 90% AL, 10% PU ALLOY

## COOLING CURVES



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## X-RAY DIFFRACTION STUDIES

Specimens for securing Debye patterns were made by filing the equilibration heat-treated half-buttons and placing the filings in clear silica capillaries. The filled capillaries were then evacuated to a pressure of  $10^{-4}$  mm of Hg and sealed. Patterns were obtained on all specimens in the as-filed condition and after subsequent heat treatments. The Straumanis<sup>(1)</sup> technique was used with a 114.7-mm diameter powder camera for the production of the Debye patterns. To obtain precision lattice measurements, stress-relieved filings were mounted between layers of Scotch Tape and exposed in a back-reflection focussing camera. Resulting patterns were analyzed by Cohen's<sup>(2)</sup> method.

In order to determine the solid solubility of plutonium in aluminum, lattice-constant measurements were made of the aluminum-rich phase in 99, 98 and 95 atomic percent aluminum alloys. As a check, measurements were made of the lattice constant of the pure aluminum melting stock. The following results were obtained.

- 
- (1) M. Straumanis and A. Ievins, Naturwissenschaften, Vol. 23, p. 833 (1935).
- (2) M. U. Cohen, Rev. Sci. Inst., Vol. 6, p. 68 (1935) and Vol. 7, p. 155 (1936).

| Nominal<br>Composition<br>(At.% Al) | H. T. of<br>Original<br>Specimen | H.T. of<br>Filings for<br>Stress Relief | $a_0$ at 25°C(kX)   |                     |
|-------------------------------------|----------------------------------|---|---------------------|---------------------|
|                                     |                                  |   | 1st                 | 2nd                 |
| 100                                 | ----                             | 150°C-15 hr.-A.C.                       | 4.0414 <sub>4</sub> | 4.0413 <sub>9</sub> |
| 100                                 | ----                             | 200°C-15 hr.-A.C.                       | 4.0414 <sub>8</sub> | 4.0416 <sub>2</sub> |
| 99                                  | 500°C-200 hr.-W.Q.               | 200°C-17 hr.-A.C.                       | 4.0420              | 4.0421              |
| 98                                  | 600°C-200 hr.-W.Q.               | 200°C-15 hr.-A.C.                       | 4.0418 <sub>6</sub> | 4.0416 <sub>7</sub> |
| 95                                  | 500°C-200 hr.-W.Q.               | 150°C-15 hr.-A.C.                       | 4.0414 <sub>8</sub> | 4.0419 <sub>9</sub> |

(W. Q. = water-quenched)                      (A.C. = air cooled)

The values obtained indicate that the solid solubility of plutonium in aluminum, if not zero, is too low to be detected by the lattice-constant method.

The results obtained from the various Debye patterns are summarized in Table II. Because of uncertainties of composition resulting from the relatively high vapor pressure of aluminum at the casting temperatures, spattering during melting, and gross segregation, the compositions of the intermediate phases, with one exception, have not yet been accurately determined. The composition of the phase  $\text{PuAl}_2$  was fixed by determination of its relatively simple crystal structure. In the vicinities of the compositions  $\text{PuAl}_3$  and  $\text{PuAl}_4$ , two intermediate phases of complex structure exist. Solution of these structures has not yet been accomplished. The intermediate phase  $\text{Pu}_3\text{Al}$  has proved to be difficult to

obtain in quantities large enough to yield a good diffraction pattern. However, the few lines obtained suggest a tetragonal structure.

The crystal structure of  $\text{PuAl}_2$  was determined through its isomorphism with  $\text{UAl}_2$  and  $\text{Cu}_2\text{Mg}$ . Independent intensity checks made on  $\text{PuAl}_2$  corroborated the results of Rundle (CT-2721) for  $\text{UAl}_2$ . The unit cell edge of the face-centered cubic  $\text{PuAl}_2$  was found to be 7.820 kX. The interatomic distances are Pu-Pu, 3.38 kX; Al-Pu, 3.24 kX, and Al-Al, 2.76 kX. The calculated density is 8.09 grams per cubic centimeter (observed density - 7.6 g/cc). The determinations were made on patterns obtained from a specimen which had been heat treated 200 hours at 500°C and was water-quenched.

Photograms obtained from specimens containing 25 to 50 atomic per cent aluminum after water quenching from a 200 hour - 500°C heat treatment showed the presence of delta solid solution plus  $\text{PuAl}_2$ . As a result, the existence of a broad two-phase field (delta solid solution plus  $\text{PuAl}_2$ ) was postulated. However, after the same specimens had been given an additional heat treatment at 300°C for nineteen hours (followed by cooling in air) their patterns included faint lines which were found to correspond to those of a distorted cubic structure, possibly tetragonal with an axial ratio of approximately 0.98. The conclusion may be drawn

that an intermediate phase exists in the composition region of PuAl, but the exact mechanism of its formation is unknown. Its crystal structure, combined with the fact that it becomes plastic at elevated temperatures, suggests that it may possess a fairly wide homogeneity range.

Examination of the specimens containing 20 and 25 per cent aluminum quenched from elevated temperatures revealed the presence of an additional intermediate phase. Although only a few lines could be obtained, the pattern suggested a tetragonal structure,  $\text{Pu}_3\text{Al}$ , isomorphous with  $\text{SrPb}_3^{(3)}$ . Patterns obtained from specimens heat treated at various temperatures reveal the fact that the intermediate phase  $\text{Pu}_3\text{Al}$  is present at  $550^\circ\text{C}$  but absent at  $575^\circ\text{C}$ . The pattern of the PuAl intermediate phase is present at  $575^\circ\text{C}$  but absent at  $600^\circ\text{C}$ . The sluggishness of the reactions producing  $\text{Pu}_3\text{Al}$  and PuAl, and the microstructures of such alloys, suggest that these phases may be formed by peritectoid reactions.

Precision measurements of the lattice constants of plutonium-rich alloys quenched from  $300^\circ\text{C}$  and  $500^\circ\text{C}$  indicate that the limit of solid solubility of aluminum in plutonium (delta phase) is about 12.5 atomic per cent

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(3) Zintl and Neumayr, Zeitschrift fur Electrochemie, Vol. 39, p. 86 (1933).

aluminum at these temperatures. The alpha-phase plutonium pattern was observed in the photographs of the 1 and 2 atomic percent aluminum alloys but was absent in those obtained from the 5 atomic percent alloy.

Representative Debye patterns obtained are shown in Figure 9. For those phases for which they have thus far been determined, interplanar spacings and estimated relative intensities of Debye pattern lines are given in Appendix II.

TABLE II

PHASES PRESENT IN PLUTONIUM-ALUMINUM ALLOYS  
AS DETERMINED FROM DEBYE PATTERNS

| Nominal<br>Composition<br>(At. % Al) | Treatment  | Phases Represented<br>in Debye Pattern                                       |
|--------------------------------------|--|--|
| 99                                   | Cast button-500°C-<br>200 hr. Q, filings           | Al + weak PuAl <sub>4</sub>  |
| 95                                   | Same as above                                      | Al + PuAl <sub>4</sub>   |
| 85                                   | Same as above                                      | Al + PuAl <sub>4</sub>   |
| 80                                   | Same as above                                      | Very weak Al + PuAl <sub>4</sub>   |
| 75                                   | Same as above                                      | PuAl <sub>3</sub> + very weak PuAl <sub>4</sub>                              |
| 70                                   | Same as above                                      | PuAl <sub>2</sub>  |
| 50                                   | Same as above                                      | PuAl <sub>2</sub> plus delta<br>solid solution                               |
| 50                                   | Same as above plus<br>300°C-165 hr. - Q            | PuAl <sub>2</sub> plus delta solid<br>solution plus medium PuAl              |
| 40                                   | Cast button-500°C-<br>200 hr. - Q, filings         | PuAl <sub>2</sub> plus delta<br>solid solution                               |
| 40                                   | Same as above plus<br>300°C-165 hr.                | PuAl <sub>2</sub> plus delta solid<br>solution plus med. PuAl                |
| 25                                   | Cast button-500°C-<br>200 hr. - Q, filings         | Delta solid solution<br>plus weak PuAl <sub>2</sub>                          |
| 25                                   | Same as above plus<br>300°C-19 hr. - Q             | Delta solid solution<br>plus weak PuAl <sub>2</sub> ,<br>plus very weak PuAl |
| 25                                   | Exposed in high-<br>temperature camera<br>at 450°C | Delta solid solution<br>plus weak PuAl plus<br>weak Pu <sub>3</sub> Al       |

TABLE II (Continued)

| Nominal<br>Composition<br>(At. % Al) | Treatment                                   | Phases Represented<br>in Debye Pattern                  |
|--------------------------------------|---|---|
| 10                                   | Cast button-500°C-<br>200 hr. - Q, filings  | Delta solid solution                                    |
| 10                                   | Same as above plus<br>300°C-70 hr. - Q      | Delta solid solution                                    |
| 5                                    | Cast button-500°C-<br>200 hr. - Q, filings  | Delta solid solution                                    |
| 5                                    | Same as above plus<br>300°C-165 hr. - Q     | Delta solid solution                                    |
| 2                                    | Cast button, 500°C-<br>200 hr. - Q, filings | Delta solid solution<br>plus very weak alpha            |
| 2                                    | Same as above plus<br>300°C-70 hr. - Q      | Delta solid solution                                    |
| 1                                    | Cast buttons-500°C-<br>200 hr. - Q, filings | Delta solid solution<br>plus strong alpha<br>plutonium. |
| 1                                    | Same as above plus<br>300°C-19 hr. - Q      | Delta solid solution.                                   |

FIGURE 9A. Pure aluminum (99.9968% Al)  
Filings stress-relieved at 200°C - 15 hours -  
A.C.\* Exhibits Al pattern.

FIGURE 9B. 95 atomic percent aluminum, Specimen  
Number 293. H.T. at 500°C - 238 hours - W. Q.\*\*  
Filings stress-relieved at 150°C - 70 hours - A. C.  
Exhibits Al plus PuAl<sub>4</sub> patterns.

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\*A.C. - Air-cooled in sealed capillary or capsule.

\*\*W.Q. - Water-quenched in sealed capillary or capsule.

FIGURE 9C. 80 atomic percent aluminum, Specimen Number 312. H.T. at 500°C - 238 hours - W.Q. - as filed. Exhibits one phase, PuAl<sub>4</sub> pattern.

FIGURE 9D. 75 atomic percent aluminum, Specimen Number 313. H.T. at 500°C - 238 hours - W.Q. Crushed easily. Exhibits one phase, PuAl<sub>3</sub> pattern.

FIGURE 9E. 70 atomic percent aluminum, Specimen Number 297. H.T. at 500°C - 238 hours - W.Q. As filed. Exhibits one phase, PuAl<sub>2</sub> pattern. (Composition by chemical analysis: 66 atomic percent aluminum).

FIGURE 9F. 40 atomic percent aluminum, Specimen Number 303. H.T. at 500°C - 200 hours - W.Q. Filings stress-relieved at 300°C - 165 hours -W.Q. Exhibits PuAl<sub>2</sub> plus delta plus PuAl patterns. The lines attributed to the PuAl phase are marked.

FIGURE 9G. 10 atomic percent aluminum, Specimen Number 309. Cast button cold-worked and H.T. at 500°C - 200 hours - W.Q. Filings stress-relieved at 300°C - 70 hours - W.Q. Exhibits delta-phase pattern. Note that the positions of delta-pattern lines are approximately the same as in Figure 9F.

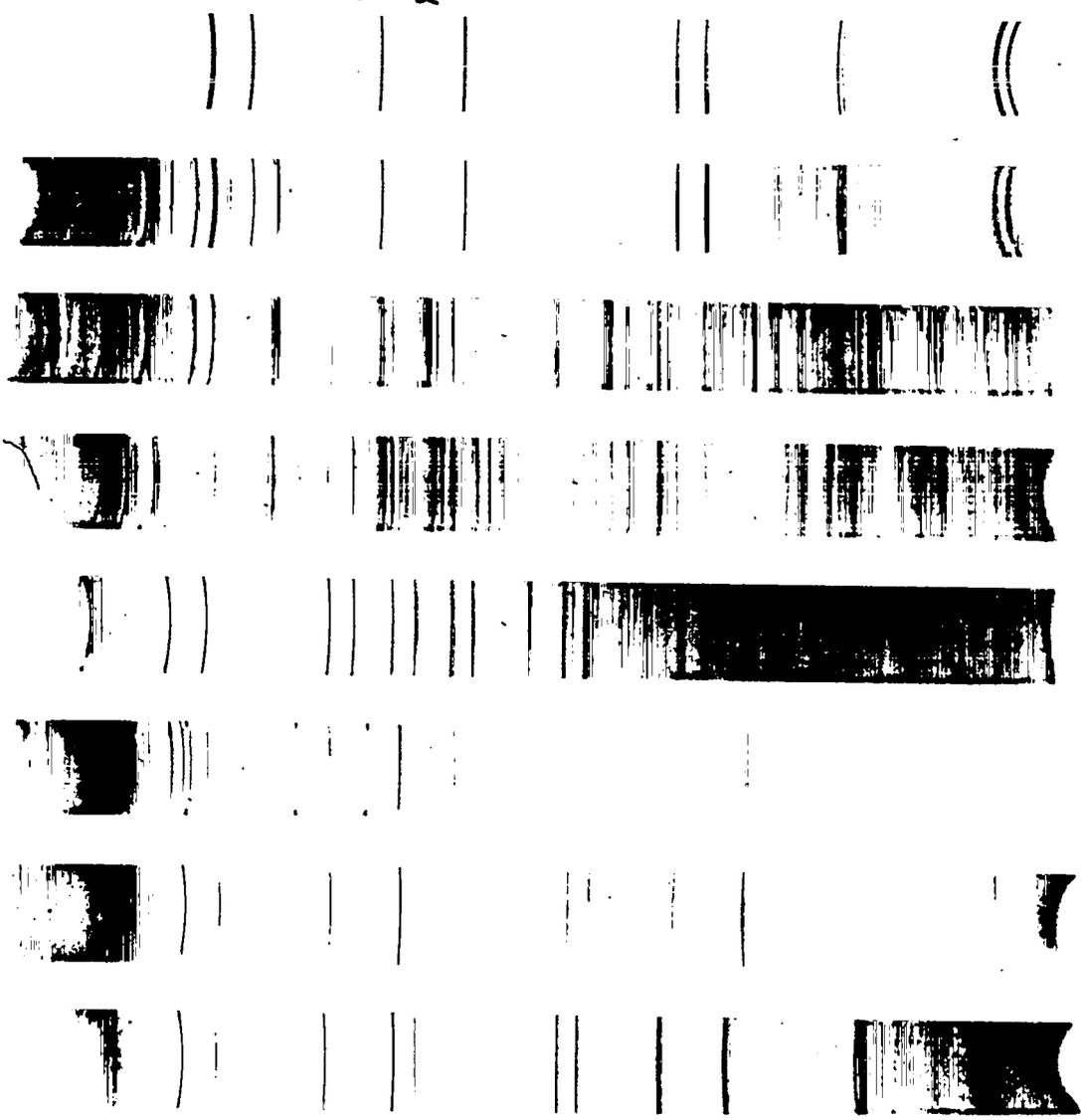
FIGURE 9H. 1 atomic percent aluminum, Specimen Number 318. Cast button cold-worked and H.T at 500°C - 200 hours - W.Q. Filings stress-relieved at 300°C - 19 hours - W.Q. Exhibits delta-phase with resolved doublets. In conjunction with Figure 9G, shows the decrease in  $a_0$  with increasing aluminum content.

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Figure 9



Cu K $\alpha$  Radiation



|   | % Al | Phases                                |
|---|------|---------------------------------------|
| A | 100  | Al                                    |
| B | 95   | Al + PuAl <sub>4</sub>                |
| C | 80   | PuAl <sub>4</sub>                     |
| D | 75   | PuAl <sub>3</sub>                     |
| E | 70   | PuAl <sub>2</sub>                     |
| F | 40   | PuAl <sub>2</sub> + PuAl + $\delta$   |
| G | 10   | $\delta$ ( $a_0 = 4.588 \text{ kX}$ ) |
| H | 1    | $\delta$ ( $a_0 = 4.624 \text{ kX}$ ) |

## MICROSTRUCTURAL INVESTIGATION

The specimens used for microscopy were mounted in prefabricated lucite mounts with methyl-methacrylate plastic and faced-off on a small lathe. The mounted specimens were then polished through 4/0 emery paper and finish-polished on a wheel using Gamal cloth and Gamal alumina. This procedure became less satisfactory as the percentage of plutonium was increased but was utilized over the entire range of alloys.

The etching procedure varied only slightly among alloys containing between 20 and 100 atomic percent aluminum, and consisted of a swab etch with a solution of 2 to 3 percent sodium hydroxide in water for periods ranging from 5 seconds to 20 seconds. For alloys containing less than 20 atomic percent aluminum, an electrolytic etch, similar to that which has been developed for pure plutonium, proved to be superior to the sodium hydroxide etch. The electrolytic etching of these alloys required variations in current density from alloy to alloy, but the electrolyte in all cases was a solution of four per cent tetraphosphoric acid and two percent glycerine in water. In several instances, an improvement in delineation of phases was obtained by following electrolytic etching with a light mechanical polish,

using Gamal alumina on a wheel covered with Gamal cloth. This variation was used successfully on all alloys containing less than about 10 atomic percent aluminum.

Specimens in both the as-cast condition and after heat treatment for 200 hours at 500°C were examined microscopically. Since the heat treatment did not appreciably alter the structure in any case, the photomicrographs presented are of alloys in the as-cast condition, except Figures 32 through 35, which for comparison show heat-treated structures.

The microstructures of the alloys showed the existence of a eutectic composition at about 98 atomic percent aluminum. The eutectic structure was shown to exist in all alloys containing more than about 80 atomic percent aluminum. Microhardness determinations indicated that the two phases of the eutectic were substantially pure aluminum and a relatively hard and brittle intermediate phase. Microstructures of alloys containing between 80 and 65 atomic percent aluminum suggested the presence of three intermediate phases containing about 80, 75, and 67 atomic percent aluminum. From microscopic examination it appeared that the region between 67 and 15 atomic percent aluminum was a broad two phase field composed of  $PuAl_2$  plus delta solid solution. Microhardness tests, however, yielded very erratic results which indicated the possible presence of one

or more additional phases. At no time were these suspected phases observed microscopically, but their postulated existence has been substantiated by the discovery, through x-ray diffraction, of the phases  $\text{PuAl}$  and  $\text{Pu}_3\text{Al}$ .

Both hardness tests and microscopic examinations have shown that, in alloys containing less than about 2 atomic percent aluminum, two phases exist at room temperature (delta solid solution plus alpha plutonium).

The structure exhibited in Figures 10 and 11 is typical of a eutectic structure plus primary phase (aluminum). The alloy shown in Figure 12 shows a nearly 100% eutectic structure, while Figure 13 exhibits a typical intermediate phase ( $\text{PuAl}_4$ ) plus eutectic structure. Figures 15 and 16 indicate the rapidly increasing amount of  $\text{PuAl}_4$  up to the 80 atomic percent aluminum alloy shown in Figures 17 and 18. Figures 19 through 23 indicate an absence of eutectics between the intermediate phases and thereby suggest peritectic reactions. Figures 24 through 29 show the steadily decreasing amount of the  $\text{PuAl}_2$ (white) intermediate phase. Figure 30 indicates that alloys containing approximately 10 atomic percent aluminum consist largely of the delta solid solution, and Figure 31 shows a structure consisting of a mixture of the delta solid solution and alpha plutonium.

After photographing, the polished and etched alloys were given microhardness tests on a Tukon Microhardness Tester using a Vickers type indenter. The results obtained exhibited considerable variation, but average values of the hardnesses of microconstituents are listed in Table III.

Lineal analyses of amounts of phases were attempted, but were found to be subject to such large errors as to make the results of no value.

TABLE III

| Microconstituent     | Nominal Composition<br>(At. % Al) | D.P.H.N.* |
|----------------------|-----------------------------------|-----------|
| Pure Aluminum        | 100                               | 17        |
| Eutectic             | 98.3                              | 48-52     |
| PuAl <sub>4</sub>    | 80.0                              | 500-525   |
| PuAl <sub>3</sub>    | 75.0                              | 400-450   |
| PuAl <sub>2</sub>    | 66.7                              | 500-600   |
| PuAl                 | 50.0                              | 100-150   |
| Pu <sub>3</sub> Al   | 25.0                              | 150-200   |
| Delta Solid Solution | 5.0-12.5                          | 95-100    |
| Alpha Plutonium      | 0.0                               | 150-175   |

\*Diamond Pyramid Hardness - 25-gram load on Vickers type indenter in Tukon Microhardness Tester.

An attempt was made to develop a convenient and reasonably rapid technique for differentiating between phases of differing plutonium content. Preliminary results were gratifying and are illustrated in Figure 36 and Figure 37. The technique, which is here called autoradiomicrography, consists of making an autoradiograph by placing the freshly polished metallographic specimen in direct contact with a photographic film of extremely fine grain, and subsequently making a photomicrograph of the developed autoradiograph.

Figure 36, the autoradiomicrograph of a 99 atomic percent aluminum alloy, can be compared with Figure 10, the photomicrograph of the same alloy. The autoradiomicrograph shows that the plutonium atoms are concentrated in the eutectic structure. Figure 37 shows the degree of segregation of a 65 atomic percent alloy and should be compared with Figure 23. There seem to be three degrees of concentration of plutonium atoms shown, perhaps the result of a sluggish reaction.

The above two autoradiomicrographs were made for Group CMR-5 by Group GMX-1.

FIGURE 10. Specimen Number 321a, 99 percent aluminum, as cast. Illustrates the aluminum- $PuAl_4$  eutectic in the interstices of the primary aluminum dendrites. Swab-etched in 2% NaOH - 15-20 seconds. Magnification 100 diameters.

FIGURE 11. Specimen Number 321a. Same as Figure 10, except magnification is 500 diameters.

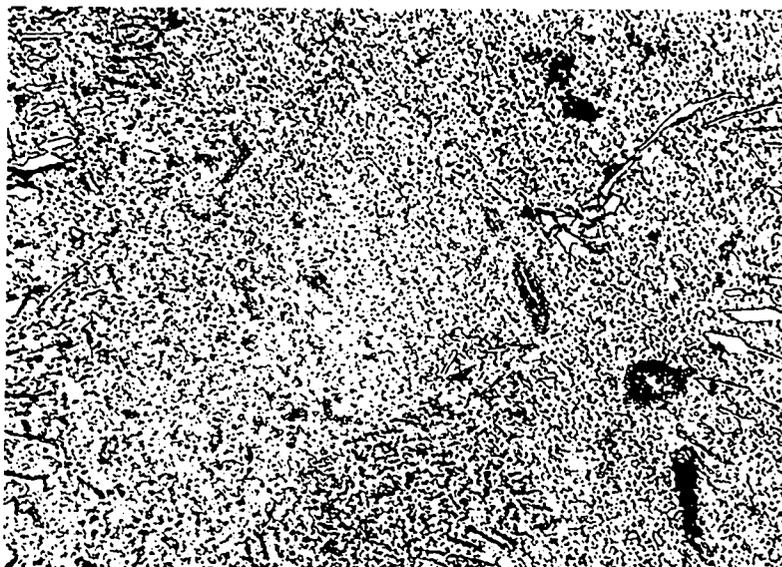
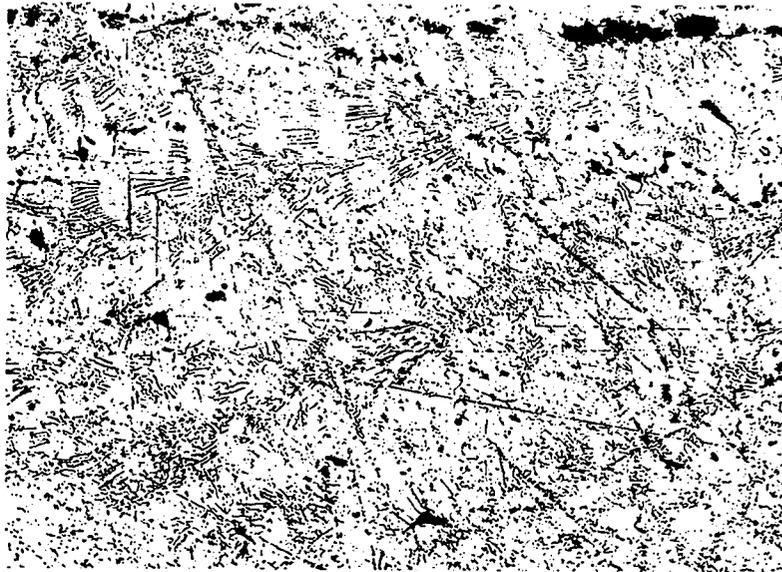


FIGURE 12. Specimen Number 292a, 98.1 percent aluminum, as cast. Almost 100% eutectic, with small excess of  $PuAl_4$ . Swab-etched in 2% NaOH 15-20 seconds. Magnification 100 diameters.

FIGURE 13. Specimen Number 293a, 95.1 percent aluminum, as cast.  $PuAl_4$  in eutectic matrix. Swab-etched in 2% NaOH - 15-20 seconds. Magnification 100 diameters.

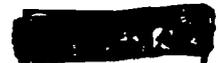
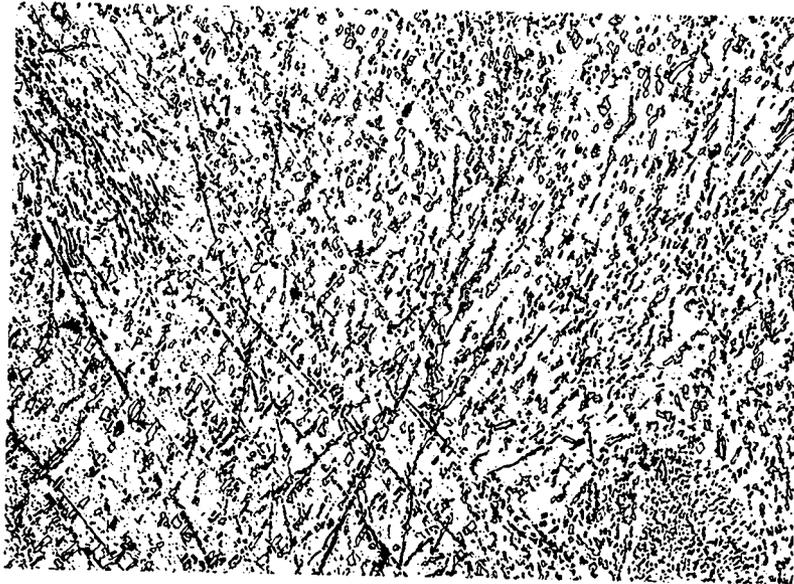


FIGURE 14. Specimen Number 95.1 percent  
aluminum, as cast. Same as Figure 13 except  
unetched. Mechanically polished.  
Magnification 100 diameters.

FIGURE 15. Specimen Number 294a, 85.6 percent  
aluminum, as cast.  $PuAl_4$  in matrix of eutectic.  
Swab-etched in 2% NaOH 15 to 20 seconds.  
Magnification 100 diameters.

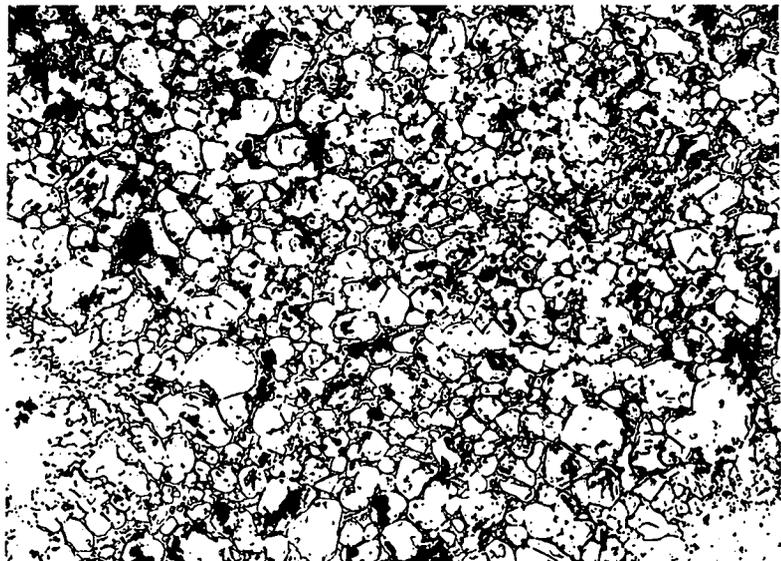
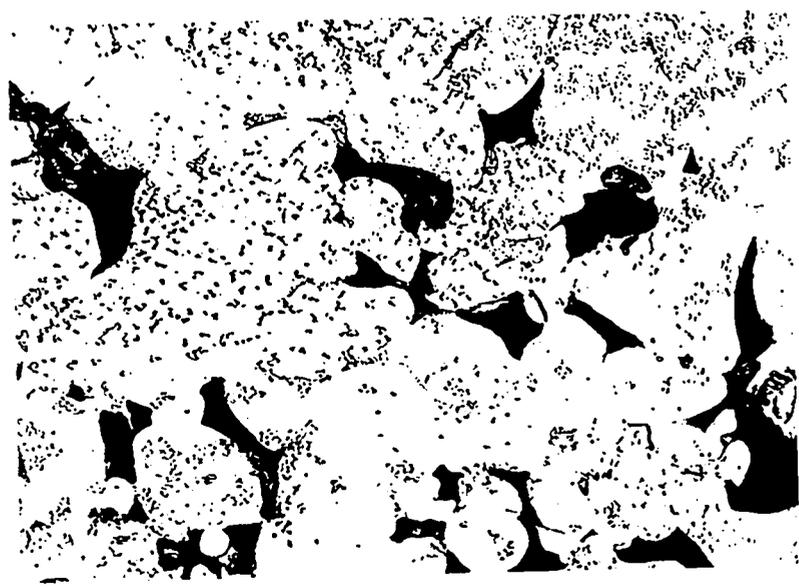
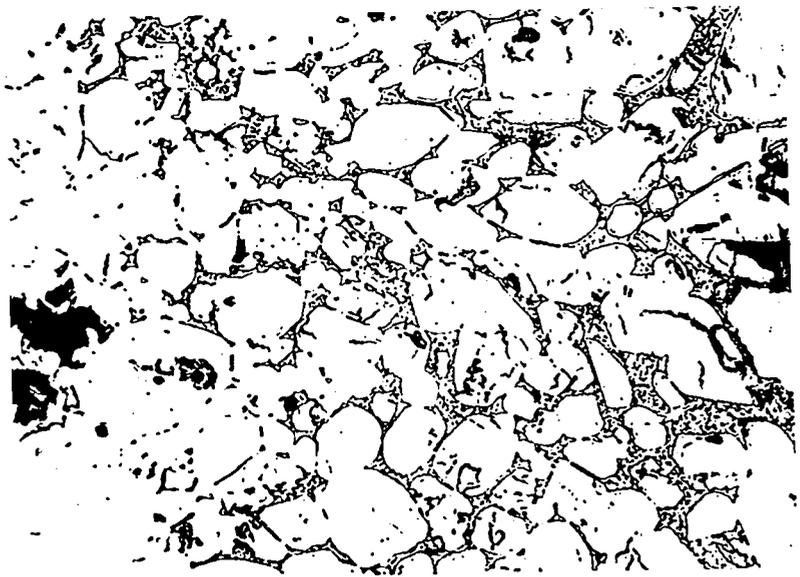




Figure 16. Specimen Number 294a, 85.6 percent aluminum, as cast. Same as Figure 15 except for higher magnification. Clearly shows the eutectic matrix. Magnification 500 diameters.

Figure 17. Specimen Number 312a, 80 percent aluminum (chemical analysis: 82.4 per cent aluminum), as cast. Almost entirely  $PuAl_4$  with little eutectic. Eutectic easily resolved under polarized light. Black portions are voids produced in casting. Swab-etched in 2% NaOH 15-20 seconds. Magnification 500 diameters.

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FIGURE 18. Specimen Number 312a, 80 percent aluminum, as cast. Same as Figure 17 except lower magnification and unetched. Does not resolve the small amount of eutectic present. Magnification 250 diameters.

FIGURE 19. Specimen Number 313a, 75 percent aluminum (chemical analysis: 77.1 per cent aluminum), as cast. Apparently the intermediate phase  $\text{PuAl}_3$ . Swab-etched in 2-3% NaOH - 5 seconds. Magnification 500 diameters.

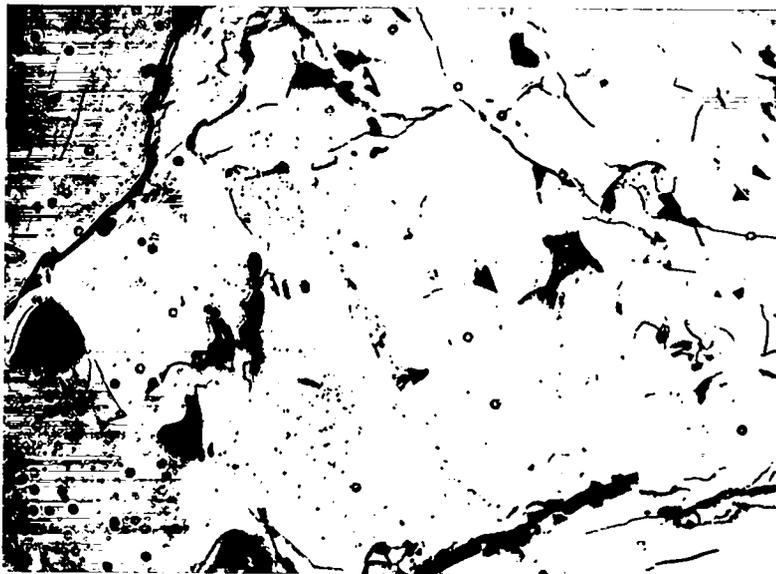


FIGURE 20. Specimen Number 314a, 69.2 percent aluminum, as cast. Two-constituent structure, both constituents appear to be compounds ( $\text{PuAl}_3$  and  $\text{PuAl}_2$ ). Swab-etched in 2 to 3 per cent NaOH - 5 seconds. Magnification 250 diameters.

FIGURE 21. Specimen Number 297a, 70 percent aluminum (chemical analysis: 66.0 per cent aluminum), as cast. A second constituent in the interstices of  $\text{PuAl}_2$  dendrites. Mechanically polished, unetched. Magnification 100 diameters.

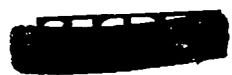
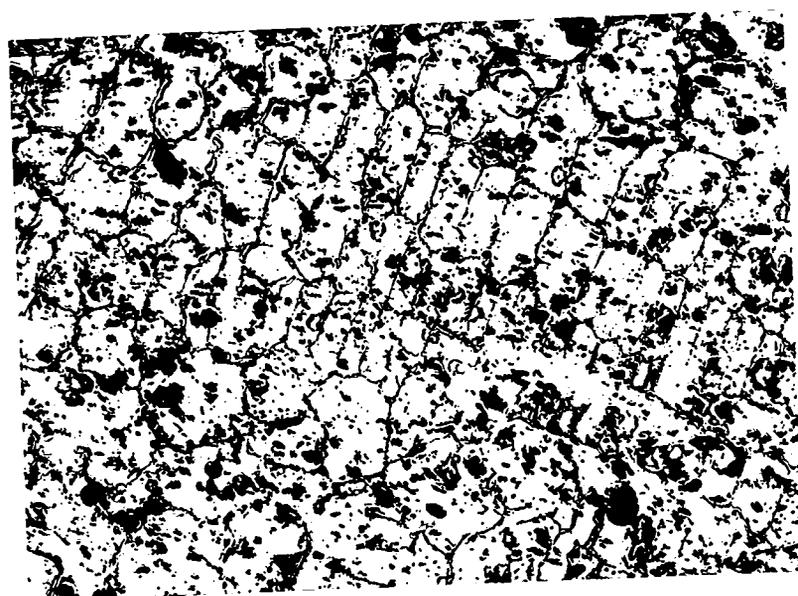


FIGURE 22. Specimen Number 297a, 70 percent aluminum, as cast. Same as Figure 21 except higher magnification. Any fine structure of the gray constituent is not resolved, either with or without etching, or with polarized light. Magnification 500 diameters.

FIGURE 23. Specimen Number 298a, 65 percent aluminum, as cast, Predominantly an intermediate phase ( $\text{PuAl}_2$ ). Swab-etched in 2-3 per cent NaOH - 5 seconds. Magnification 100 diameters.

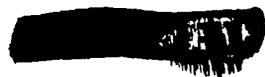
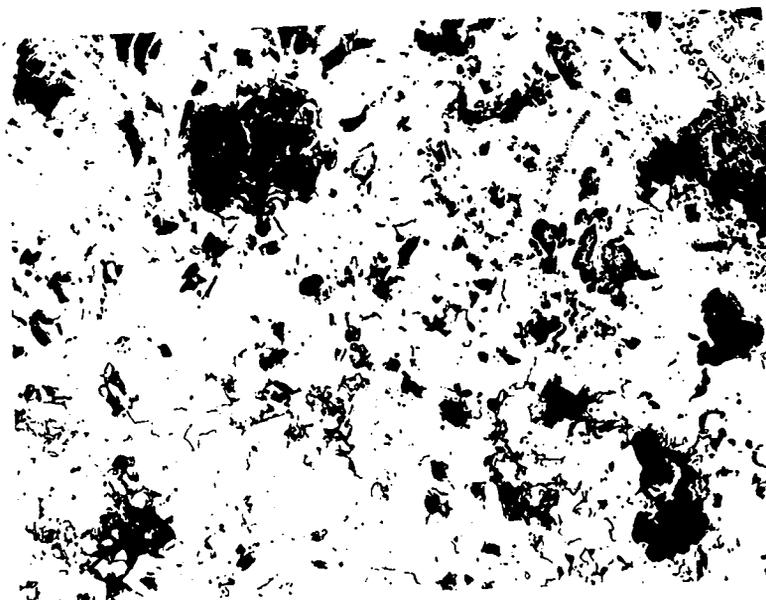
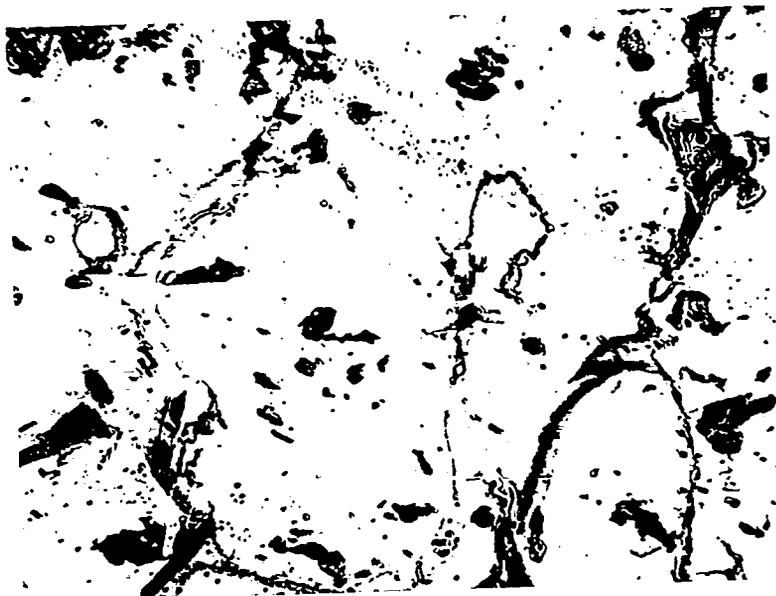


FIGURE 24. Specimen Number 299a, 58.9 percent aluminum,  
as cast. Appears to be two phases ( $\text{PuAl}_2$  and delta).  
Mechanically polished, unetched. Magnification 500 diameters.

FIGURE 25. Specimen Number 300a, 55 percent aluminum,  
as cast. Mechanically polished, unetched.  
Magnification 500 diameters.

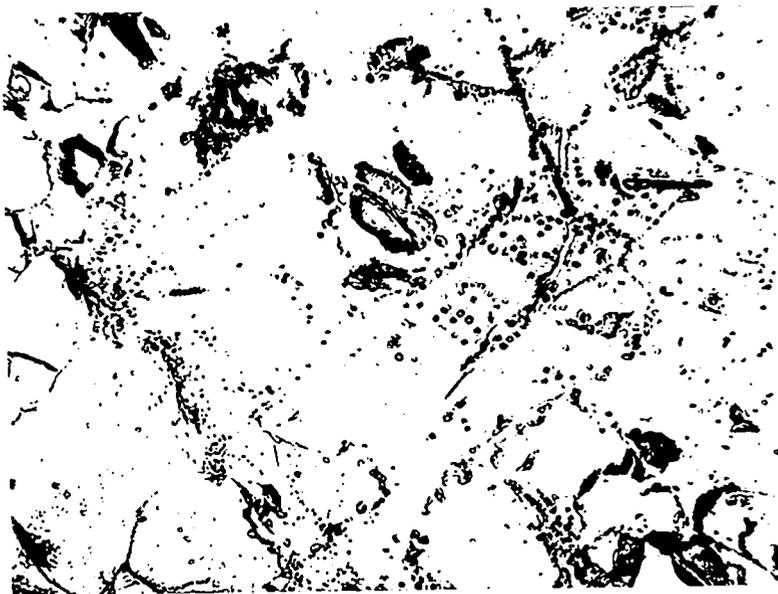


FIGURE 26. Specimen Number 302a, 45 percent aluminum,  
as cast. Mechanically polished, unetched.  
Magnification 500 diameters.

FIGURE 27. Specimen Number 304a, 35 percent aluminum,  
as cast. Swab-etched in 2% NaOH 15-20 seconds.  
Magnification 500 diameters.

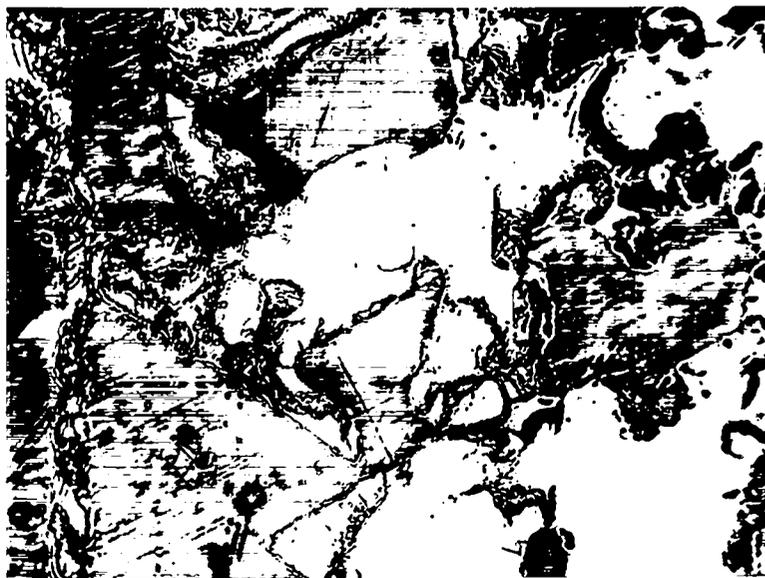


FIGURE 28. Specimen Number 305a, 30 percent aluminum,  
as cast. Mechanically polished, unetched.  
Magnification 500 diameters.

FIGURE 29. Specimen Number 307a, 20 percent aluminum,  
as cast. Mechanically polished, unetched. Magnification 500  
diameters.

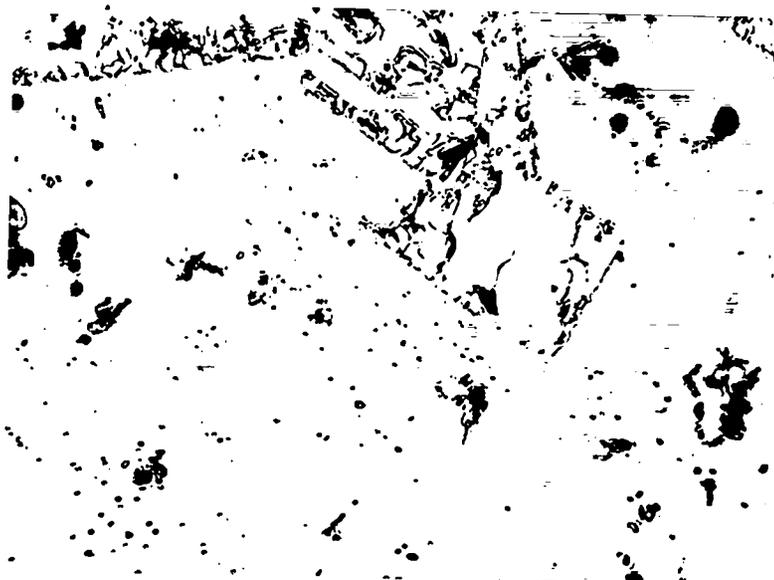
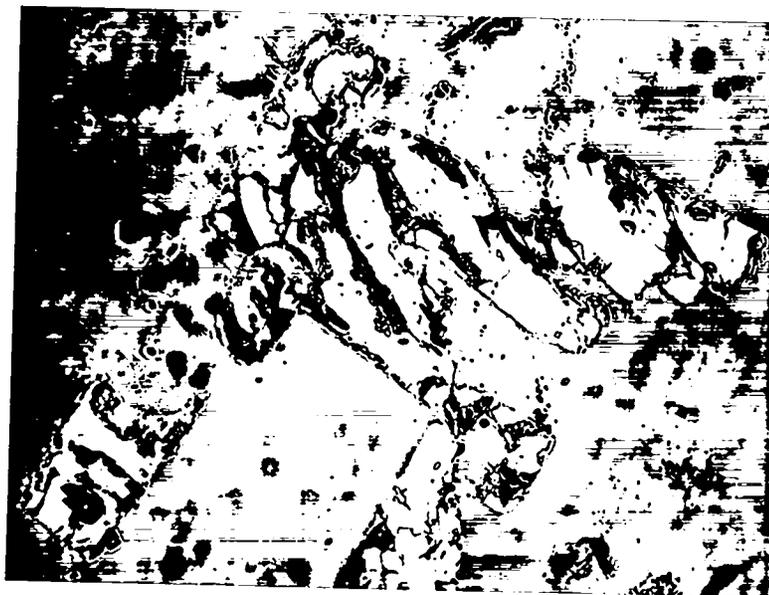


FIGURE 30. Specimen Number 309a, 10 percent aluminum, as cast, Nearly 100% delta solid solution. Mechanically polished, electrolytically etched in 4% tetraphosphoric acid, and lightly repolished mechanically. Magnification 100 diameters.

FIGURE 31. Specimen Number 318a, 1 percent aluminum, as cast. Alpha plutonium in a delta solid solution matrix. Electrolytically etched in 4% tetraphosphoric acid and lightly repolished mechanically. Magnification 100 diameters.

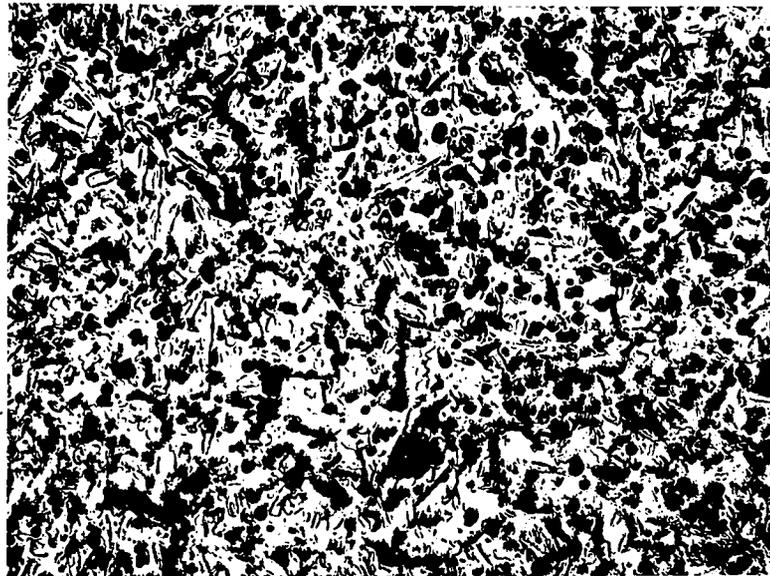
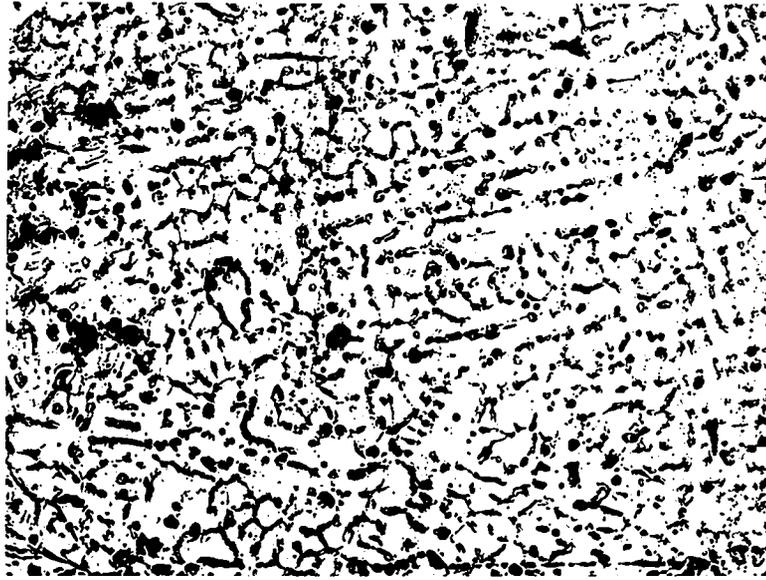
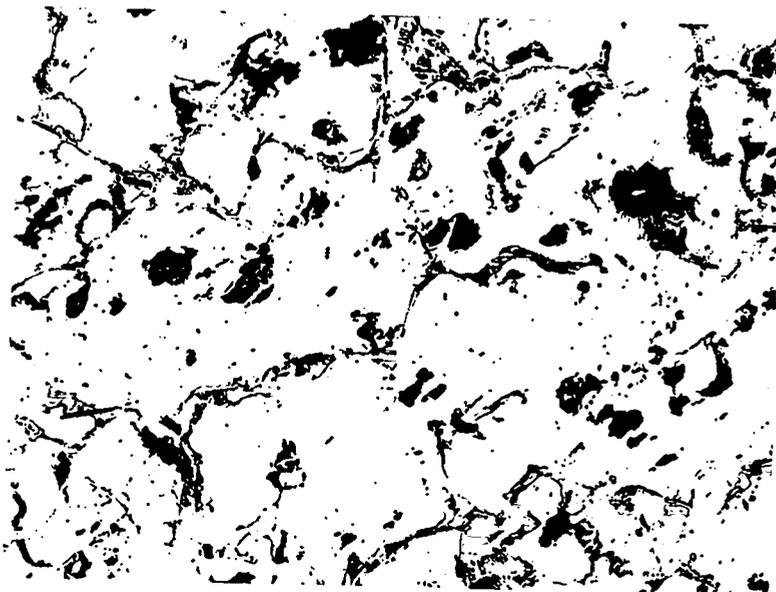


FIGURE 32. Specimen Number 292. 98.1 percent aluminum.  
As cast and equilibration heat treated for 200 hours at  
600°C and water quenched. Essentially the same as Figure  
12. Swab-etched in 2% NaOH. Magnification 250 diameters.

FIGURE 33. Specimen Number 297. 70 percent aluminum.  
As cast and equilibration heat treated for 238 hours at  
500°C and water quenched. Essentially the same as Figure  
21. Mechanically polished, unetched. Magnification 250  
diameters.

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FIGURE 34. Specimen Number 299. 58.9 percent aluminum, as cast and equilibration heat treated for 300 hours at 500°C and water quenched. Mechanically polished unetched. Compare with Figure 24. Magnification 250 diameters.

FIGURE 35. Specimen Number 338. 12.5 percent aluminum, as cast. Exhibits Vickers indentations indicating three different hardnesses. Electrolytically etched in 4% tetraphosphoric acid. Magnification 500 diameters.

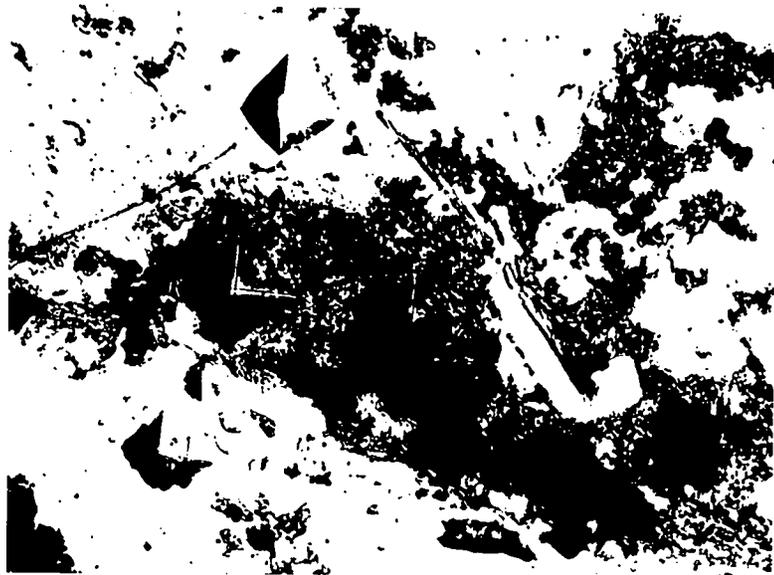
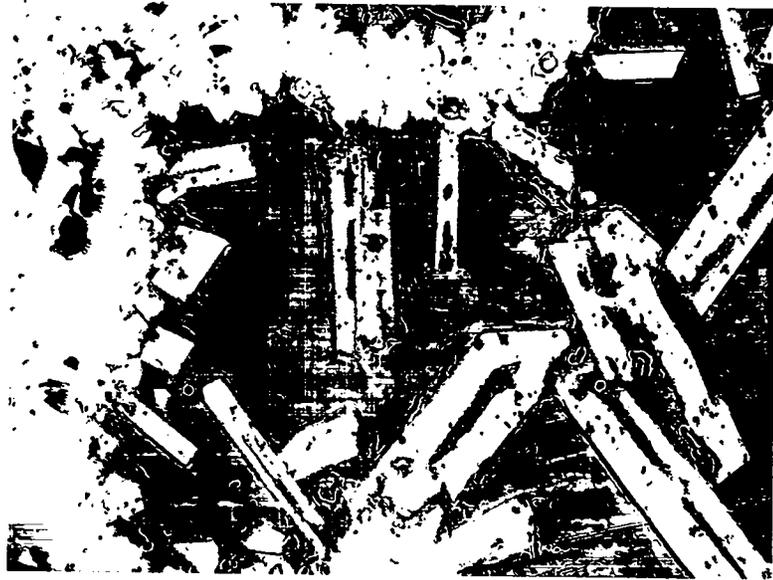


FIGURE 36. Autoradiomicrograph of Specimen Number 321a, 99 percent aluminum. Shows the concentration of plutonium in the eutectic structure. Magnification 25 diameters.

FIGURE 37. Autoradiomicrograph of Specimen Number 298a, 65 percent aluminum. Shows three levels of plutonium content, presumably because of a sluggish reaction and non-equilibrium state. Magnification 25 diameters.



## MACHINING AND PLASTIC FORMING

A half-inch diameter cast rod of 90 atomic percent aluminum alloy was found to have a machinability quite superior to that of pure aluminum. In turning this rod down to 1/4-inch diameter in a lathe, it was found to have about as good machining characteristics as gray cast iron.

Prior to equilibration heat treatments, attempts were made to hand-forge the cast alloy buttons. Those containing high percentages of delta solid solution (2 to 20 atomic percent aluminum) were found to be quite malleable and were successfully cold-hammered into rectangular bars. Alloys containing 98 and 99 atomic percent aluminum were cold-forged successfully but with some difficulty. A 98.5 atomic percent aluminum alloy was hot-extruded from a 1/2-inch cylinder to form 1/4-inch rod, but again the operation was difficult, doubtless because of the strengthening effect of the eutectic structure. Pure aluminum was extruded from the same die at a temperature of 210°C under a pressure of 30,000 lbs. per square inch; but the 98.5 percent aluminum alloy required a temperature of 425°C and a load of 45,000 pounds per square inch. A second extrusion of the same alloy performed at 485°C under a load of 50,000 pounds per square inch yielded a rod of better appearance than the

first. All three rods were prepared for use as thermal-conductivity specimens.

The two 98.5 atomic percent aluminum rods and the pure aluminum rod were formed in an indirect extrusion-type die having a tight-fitting plunger (clearance less than 0.0005 inch). A fourth 1/4-inch rod was formed from a 95 atomic percent aluminum alloy in a direct extrusion die having a plunger 0.040-inch undersize. While this resulted in the production of a 0.020-inch thickness of flash between plunger and die wall, it permitted this alloy containing approximately 28 volume percent  $PuAl_4$  to be extruded into a satisfactory thermal conductivity specimen at a temperature of 275°C, a pressure of 50,000 pounds per square inch, and a speed of 4 inches per minute (approximately ten times the maximum speed attained with pure aluminum in the indirect extrusion die).

Attempts were made to press both 80 atomic percent and 70 atomic percent alloys at 485°C. Both were definitely brittle at this temperature and showed no evidence of plastic deformation. A 45 atomic percent aluminum alloy was pressed at 450°C and, though the alloy was quite brittle at room temperature, it was found capable of undergoing plastic deformation to a considerable extent at the pressing temperature. An alloy containing 25 atomic percent

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aluminum was pressed at 430°C and evidenced definite plastic deformability at this temperature. The 25 atomic percent alloy was not so workable as the 45 percent alloy at elevated temperatures, and was just as brittle at room temperature.

The 5, 10, and 15 atomic percent aluminum alloys were rolled at room temperature with a reduction of between 75 and 90 percent. The cold workability of these alloys is excellent, though there was definite evidence of work hardening in all instances.

From these data, good cold workability can be expected between 2 and 20 atomic percent aluminum. Between 20 and 50 atomic percent, it is necessary to attain temperatures in the region of 450°C before appreciable plastic deformation can be expected. All alloys between 50 and 90 atomic percent aluminum were found to be brittle, and no plastic deformability was noted up to 485°C, the highest temperature investigated. Alloys between 90 and 100 atomic percent aluminum require temperatures above 250°C in order to be easily worked, but they are sufficiently malleable at room temperature to allow some deformation.

## CORROSION OBSERVATIONS

A systematic investigation of the corrosion characteristics of plutonium-aluminum alloys has not yet been undertaken. However, in the course of other work, preliminary observations of corrosion behavior have been made. It has been found that the polished surfaces of metallographic specimens containing more than about 75 atomic percent aluminum will remain virtually untarnished for periods of several weeks in laboratory atmosphere at room temperature. With decreasing aluminum content, an increasing rate of oxidation was observed, but this did not seem excessive in any instance.

## THERMAL CONDUCTIVITIES

Although results of experimental measurements are not yet available\*, a few deductions may be made regarding thermal conductivities to be expected for some plutonium-aluminum compositions. The rule of mixtures should apply to the two-phase alloys containing from 80 to 100 atomic percent aluminum. For example, since the solid solubility of plutonium in aluminum has been found to be essentially zero, the thermal conductivity of an alloy containing 91 atomic percent aluminum (50 volume percent  $\text{PuAl}_4$ , 47 weight percent plutonium) may be expected to be about half that of pure aluminum, even if the thermal conductivity of  $\text{PuAl}_4$  is assumed to be zero.

The delta solid solutions, containing from 2 to 12.5 atomic percent aluminum, have the same close-packed cubic crystal structure as the delta phase of pure plutonium; and throughout their entire range of aluminum content they closely resemble delta plutonium in all such "good metallic" properties as bright metallic luster and good plastic

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\*Since this report was written, preliminary measurements of thermal conductivities have indicated that (1) the rule of mixtures is closely approximated for alloys in the 90 to 100 atomic percent aluminum range, and (2) the thermal conductivity of the aluminum phase in these alloys must be substantially the same as that of pure aluminum.

deformability. It seems probable, therefore, that their thermal conductivities may be the same order of magnitude as that of delta plutonium, which according to unpublished data is about  $0.02 \text{ cal/cm}^2 \text{ sec/}^\circ\text{C/cm}$  at room temperature (approximately one-third the thermal conductivity of stainless steel). Compositions from 12.5 to about 42 atomic percent aluminum, as cast, would consist of  $\text{PuAl}_2$  in 50 or more volume percent of a delta-phase matrix, and hence by the rule of mixtures should have at least half the thermal conductivity of the 12.5 atomic percent alloy. However, these compositions (12.5 to 42 percent aluminum) may not be stable against thermal cycling (see discussions of  $\text{Pu}_3\text{Al}$  and  $\text{PuAl}$  above).

It appears, therefore, that alloys containing from about 25 to 85 atomic percent aluminum would be composed largely of brittle intermetallic compounds and are almost certain to be poor thermal conductors. While powdered  $\text{PuAl}_2$  could probably be sintered into compacts with powdered aluminum as a matrix to improve conductivity, the incompatibility of these phases makes it seem likely that, even at only moderately high temperatures, loss of the aluminum matrix by solid state reaction would occur.

## SUMMARY

The data, deductions, and speculations set forth in this report are presented graphically as a tentative phase diagram in Figure 38.

Data which can be considered as correct within the degree of error of the investigative methods utilized fix the following features of the diagram:

1. Essentially zero solid solubility of plutonium in aluminum at all temperatures.

2. A eutectic reaction at 98.3 atomic percent aluminum and 647°C.

3. An intermediate phase of composition  $\text{PuAl}_2$  having the face-centered cubic crystal structure of  $\text{Cu}_2\text{Mg}$  and  $a_0$  equal to 7.280 kX.

4. A face-centered cubic solid solution of aluminum in delta-phase plutonium containing a maximum of 12.5 atomic percent aluminum within the temperature range 300°C to 500°C.

Data of reasonable reliability, but which will be more positively established by further work, postulate the following:

1. Existence of an intermediate phase of complex crystal structure in the composition region  $\text{PuAl}_4$ .

2. Existence of an intermediate phase of complex crystal structure in the composition region  $\text{PuAl}_3$ .

3. Existence of an intermediate phase of tetragonal crystal structure in the composition region  $\text{PuAl}$ .
4. Existence of an intermediate phase with a possibly tetragonal structure in the composition region  $\text{Pu}_3\text{Al}$ .
5. Zero solid solubility of aluminum in the alpha, beta, and gamma phases of plutonium.
6. A degree of similarity between the plutonium-aluminum and uranium-aluminum phase diagrams.
7. A degree of similarity between the plutonium-aluminum and the rare earth-aluminum phase diagram.

Speculations based on incomplete data concern the following:

1. Existence of a narrow homogeneity range for the phase  $\text{PuAl}_4$  at elevated temperatures.
2. Existence of a peritectoid reaction, delta solid solution plus  $\text{PuAl}_2$  to form  $\text{PuAl}$ , at  $585^\circ\text{C}$ .
3. Existence of a peritectoid reaction, delta solid solution plus  $\text{PuAl}$  to form  $\text{Pu}_3\text{Al}$ , at  $560^\circ\text{C}$ .
4. Existence of a peritectic reaction, liquid plus  $\text{PuAl}_3$  to form  $\text{PuAl}_4$ , at  $652^\circ\text{C}$ .
5. Existence of a peritectic reaction, liquid plus  $\text{PuAl}_2$  to form  $\text{PuAl}_3$ , at  $725^\circ\text{C}$ .
6. Estimation of the melting point of  $\text{PuAl}_2$  at about  $1250^\circ\text{C}$ .

The delta-phase solid solution is highly workable, both hot and cold, and has good casting properties. Alloys containing large amounts of the intermediate phases  $Pu_3Al$  and  $PuAl$  are brittle at room temperature but can be hot-worked at about  $450^{\circ}C$ . Alloys containing high percentages of the phases  $PuAl_2$ ,  $PuAl_3$  and  $PuAl_4$  are brittle at room temperature and exhibit no plasticity at or below  $486^{\circ}C$ , the maximum heating temperature utilized. Castabilities of alloys containing from 45 to 90 atomic percent aluminum are poor. Alloys containing from 90 to 100 percent aluminum can be easily cast, but care must be taken to avoid shrinkage cavities. These alloys can be cold-worked to only a slight degree, but can be hot-formed at temperatures above  $250^{\circ}C$ . The 90 to 100 percent compositions have a machinability superior to that of pure aluminum.

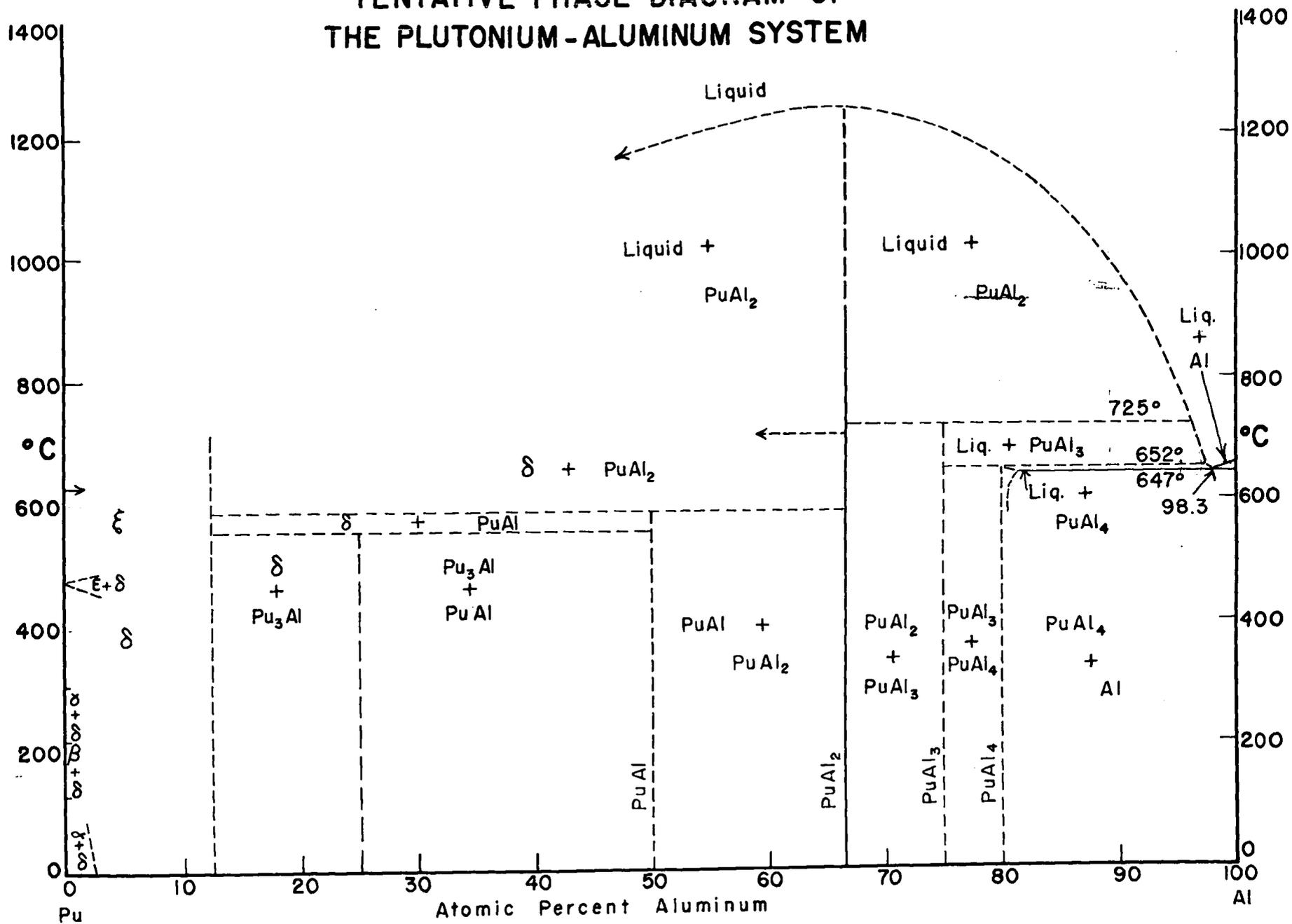
Visual observations indicate that plutonium-aluminum alloys are reasonably resistant to atmospheric corrosion in all compositions and that the alloys containing more than 75 atomic percent aluminum possess such resistance to a high degree.

It seems reasonable to expect that, with respect to order of magnitude of thermal conductivities, 2 to 20 atomic percent aluminum alloys will resemble stainless steel, 25 to 85 atomic percent aluminum alloys will be characteristic

of brittle intermetallic phases, and 90 to 100 atomic percent compositions will not be greatly different from pure aluminum.

Figure 38

# TENTATIVE PHASE DIAGRAM OF THE PLUTONIUM-ALUMINUM SYSTEM



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## APPENDIX I

## ANALYSES OF PLUTONIUM AND ALUMINUM MELTING STOCKS

Spectrochemical Analysis of Plutonium Remelt Stock RZ-16  
as Determined by Group CMR-1

| <u>Element</u> | <u>ppm</u> | <u>Element</u> | <u>ppm</u> |
|----------------|------------|----------------|------------|
| C              | 120        | Cr             | 2          |
| Na             | 6          | Mn             | 50         |
| Mg             | 14         | Fe             | 61         |
| Al             | 8          | Ni             | 20         |
| Si             | 15         | Cu             | 15         |
| Ca             | 3          | Mo             | ND*        |
| Ti             | ND*        | Ag             | 1          |
| V              | ND*        | La             | 16         |
|                |            | Pb             | 15         |

Chemical Analysis of High-Purity Aluminum - Lot Number  
5-94007-2, as Reported by the Aluminum Company of America

| <u>Element</u> | <u>Weight %</u> |
|----------------|-----------------|
| Al             | 99.9968         |
| Si             | 0.0011          |
| Fe             | 0.0006          |
| Cu             | 0.0004          |
| Mg             | 0.0007          |
| Na             | 0.0004          |

\*Not detected.

## APPENDIX II

## DEBYE PATTERN DATA FOR THE INTERMEDIATE PHASES OF THE PLUTONIUM-ALUMINUM SYSTEM

PuAl<sub>2</sub>; a<sub>0</sub> = 7.820kXCu-K-Alpha Radiation

| Miller Indices            | Interplanar Spacing | Sin <sup>2</sup> $\theta$ | Intensity |
|---------------------------|---------------------|---------------------------|-----------|
| 111                       | 4.450               | 0.0299                    | HW        |
| 220                       | 2.740               | 0.0789                    | MS        |
| 311                       | 2.341               | 0.1080                    | MS        |
| 400                       | 1.944               | 0.1567                    | HW        |
| 331                       | 1.786               | 0.1856                    | HW        |
| 422                       | 1.590               | 0.2342                    | M         |
| 511,333                   | 1.500               | 0.2633                    | M         |
| 440                       | 1.379               | 0.3115                    | HW        |
| 531                       | 1.318               | 0.3409                    | HW        |
| 620 <sub>a1</sub>         | 1.231               | 0.3897                    | HW        |
| 533 <sub>a1</sub>         | 1.189               | 0.4179                    | HW        |
| 444 <sub>a1</sub>         | 1.124               | 0.4678                    | W         |
| 711,511 <sub>a1</sub>     | 1.093               | 0.4949                    | HW        |
| 642 <sub>a1</sub>         | 1.044               | 0.5424                    | M         |
| 731,553 <sub>a1</sub>     | 1.016               | 0.5719                    | M         |
| 800 <sub>a1</sub>         | 0.976               | 0.6199                    | W         |
| 733 <sub>a1</sub>         | 0.954               | 0.6489                    | W         |
| 822,660 <sub>a1</sub>     | 0.921               | 0.6972                    | M         |
| 751,555 <sub>a1</sub>     | 0.902               | 0.7257                    | M         |
| 840 <sub>a1</sub>         | 9.874               | 0.7744                    | HW        |
| 911,753 <sub>a1</sub>     | 0.858               | 0.8036                    | M         |
| 911,753 <sub>a2</sub>     | 0.858               | 0.8071                    | HW        |
| 664 <sub>a1</sub>         | 0.833               | 0.8516                    | M         |
| 664 <sub>a2</sub>         | 0.833               | 0.9560                    | HW        |
| 931 <sub>a1</sub>         | 0.819               | 0.8799                    | M         |
| 931 <sub>a2</sub>         | 0.819               | 0.8847                    | M         |
| 844 <sub>a1</sub>         | 0.798               | 0.9279                    | MS        |
| 844 <sub>a2</sub>         | 0.798               | 0.9328                    | M         |
| 933,771,755 <sub>a1</sub> | 0.786               | 0.9568                    | MS        |
| 933,771,755 <sub>a2</sub> | 0.786               | 0.9620                    | M         |



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 PuAd<sub>3</sub> (Continued)

|    |       |        |     |
|----|-------|--------|-----|
| 40 | 1.114 | 0.4770 | HW  |
| 41 | 1.106 | 0.4844 | VW  |
| 42 | 1.082 | 0.5054 | VVW |
| 43 | 1.076 | 0.5115 | W   |
| 44 | 1.061 | 0.5259 | VW  |
| 45 | 1.039 | 0.5490 | VVW |
| 46 | 1.034 | 0.5537 | HW  |
| 47 | 1.024 | 0.5650 | W   |
| 48 | 1.014 | 0.5754 | W   |
| 49 | 1.001 | 0.5913 | VVW |
| 50 | 0.991 | 0.6033 | M   |
| 51 | 0.984 | 0.6114 | W   |
| 52 | 0.973 | 0.6254 | M   |
| 53 | 0.968 | 0.6317 | VW  |
| 54 | 0.963 | 0.6384 | VVW |
| 55 | 0.958 | 0.6452 | HW  |
| 56 | 0.942 | 0.6671 | M   |
| 57 | 0.933 | 0.6806 | HW  |
| 58 | 0.926 | 0.6912 | HW  |
| 59 | 0.914 | 0.7092 | HW  |
| 60 | 0.903 | 0.7269 | W   |
| 61 | 0.895 | 0.7388 | W   |
| 62 | 0.888 | 0.7514 | W   |
| 63 | 0.883 | 0.7596 | W   |
| 64 | 0.877 | 0.7707 | W   |
| 65 | 0.871 | 0.7809 | W   |
| 66 | 0.866 | 0.7903 | W   |
| 67 | 0.859 | 0.8029 | W   |
| 68 | 0.852 | 0.8156 | HW  |
| 69 | 0.850 | 0.8193 | W   |
| 70 | 0.854 | 0.8293 | HW  |
| 71 | 0.842 | 0.8345 | HW  |
| 72 | 0.837 | 0.8457 | VW  |
| 73 | a 1   | 0.830  | M   |
| 74 | a 2   | 0.829  | HW  |
| 75 | a 1   | 0.823  | M   |
| 76 | a 2   | 0.822  | HW  |
| 77 | a 1   | 0.818  | M   |
| 78 | a 2   | 0.818  | HW  |
| 79 |       | 0.814  | VW  |
| 80 |       | 0.812  | VW  |
| 81 |       | 0.803  | W   |
| 82 |       | 0.801  | W   |
| 83 |       | 0.798  | W   |
| 84 |       | 0.796  | M   |
| 85 |       | 0.793  | HW  |

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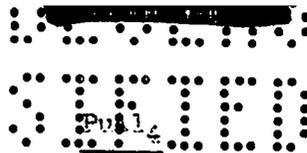
PuAl<sub>3</sub> (Continued)

|    |       |        |    |
|----|-------|--------|----|
| 86 | 0.791 | 0.9455 | HW |
| 87 | 0.789 | 0.9622 | VW |
| 88 | 0.784 | 0.9641 | HW |
| 89 | 0.782 | 0.9690 | W  |
| 90 | 0.780 | 0.9733 | W  |
| 91 | 0.777 | 0.9817 | HW |
| 92 | 0.775 | 0.9864 | M  |

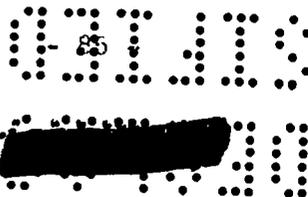
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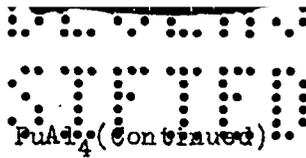
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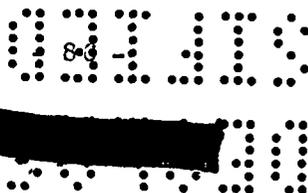
Cu-K-Alpha Radiation

| Line Number | Interplanar Spacing | $\text{Sin}^2 \theta$ | Intensity |
|-------------|---------------------|-----------------------|-----------|
| 1           | 5.576               | 0.0190                | HW        |
| 2           | 4.122               | 0.0349                | M         |
| 3           | 3.646               | 0.0446                | M         |
| 4           | 3.380               | 0.0518                | M         |
| 5           | 3.145               | 0.0599                | MS        |
| 6           | 3.102               | 0.0615                | VW        |
| 7           | 2.485               | 0.0959                | MS        |
| 8           | 2.301               | 0.1119                | MS        |
| 9           | 2.211               | 0.1211                | HW        |
| 10          | 2.178               | 0.1249                | HW        |
| 11          | 2.050               | 0.1409                | VW        |
| 12          | 2.035               | 0.1430                | W         |
| 13          | 1.917               | 0.1612                | HW        |
| 14          | 1.876               | 0.1683                | HW        |
| 15          | 1.858               | 0.1716                | HW        |
| 16          | 1.835               | 0.1759                | HW        |
| 17          | 1.806               | 0.1816                | W         |
| 18          | 1.786               | 0.1856                | VW        |
| 19          | 1.704               | 0.2040                | VW        |
| 20          | 1.645               | 0.2189                | W         |
| 21          | 1.584               | 0.2361                | M         |
| 22          | 1.558               | 0.2440                | VW        |
| 23          | 1.537               | 0.2508                | VW        |
| 24          | 1.496               | 0.2645                | HW        |
| 25          | 1.461               | 0.2773                | VW        |
| 26          | 1.448               | 0.2824                | W         |
| 27          | 1.432               | 0.2887                | W         |
| 28          | 1.418               | 0.2947                | W         |
| 29          | 1.404               | 0.3003                | VW        |
| 30          | 1.389               | 0.3071                | W         |
| 31          | 1.365               | 0.3180                | VW        |
| 32          | 1.345               | 0.3274                | VW        |
| 33          | 1.315               | 0.3423                | W         |
| 34          | 1.303               | 0.3489                | W         |
| 35          | 1.293               | 0.3543                | W         |
| 36          | 1.286               | 0.3581                | W         |
| 37          | 1.270               | 0.3669                | W         |
| 38          | 1.237               | 0.3872                | W         |
| 39          | 1.217               | 0.3996                | VW        |
| 40          | 1.205               | 0.4077                | W         |




  
 PuAd (Continued)

|    |       |        |     |
|----|-------|--------|-----|
| 41 | 1.191 | 0.4172 | HW  |
| 42 | 1.184 | 0.4224 | VVW |
| 43 | 1.177 | 0.4271 | VW  |
| 44 | 1.159 | 0.4410 | VW  |
| 45 | 1.151 | 0.4466 | VVW |
| 46 | 1.106 | 0.4841 | VVW |
| 47 | 1.095 | 0.4937 | VW  |
| 48 | 1.086 | 0.5020 | VW  |
| 49 | 1.065 | 0.5216 | W   |
| 50 | 1.058 | 0.5295 | W   |
| 51 | 1.042 | 0.5452 | VVW |
| 52 | 1.031 | 0.5573 | VVW |
| 53 | 1.021 | 0.5677 | VW  |
| 54 | 1.011 | 0.5789 | VW  |
| 55 | 0.998 | 0.5944 | HW  |
| 56 | 0.995 | 0.5978 | HW  |
| 57 | 0.990 | 0.6038 | HW  |
| 58 | 0.988 | 0.6064 | VVW |
| 59 | 0.973 | 0.6255 | W   |
| 60 | 0.960 | 0.6432 | W   |
| 61 | 0.951 | 0.6553 | W   |
| 62 | 0.946 | 0.6623 | VW  |
| 63 | 0.942 | 0.6677 | W   |
| 64 | 0.933 | 0.6807 | W   |
| 65 | 0.923 | 0.6953 | VVW |
| 66 | 0.908 | 0.7184 | VW  |
| 67 | 0.901 | 0.7289 | VW  |
| 68 | 0.898 | 0.7343 | VW  |
| 69 | 0.894 | 0.7413 | VVW |
| 70 | 0.891 | 0.7462 | VW  |
| 71 | 0.886 | 0.7549 | VW  |
| 72 | 0.883 | 0.7586 | VVW |
| 73 | 0.875 | 0.7742 | VW  |
| 74 | 0.869 | 0.7843 | W   |
| 75 | 0.867 | 0.7882 | VVW |
| 76 | 0.862 | 0.7964 | VW  |
| 77 | 0.859 | 0.8017 | VW  |
| 78 | 0.857 | 0.8068 | W   |
| 79 | 0.855 | 0.8106 | VW  |
| 80 | 0.848 | 0.8238 | VW  |
| 81 | 0.844 | 0.8311 | W   |
| 82 | 0.836 | 0.8471 | HW  |
| 83 | 0.833 | 0.8524 | HW  |
| 84 | 0.831 | 0.8576 | W   |
| 85 | 0.829 | 0.8619 | W   |


  
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PuAl<sub>4</sub> (Continued)

|     |       |        |    |
|-----|-------|--------|----|
| 86  | 0.826 | 0.8684 | M  |
| 87  | 0.819 | 0.8834 | W  |
| 88  | 0.816 | 0.8884 | HW |
| 89  | 0.812 | 0.8973 | HW |
| 90  | 0.810 | 0.9020 | VW |
| 91  | 0.808 | 0.9077 | VW |
| 92  | 0.804 | 0.9166 | VW |
| 93  | 0.801 | 0.9223 | VW |
| 94  | 0.796 | 0.9349 | W  |
| 95  | 0.793 | 0.9416 | W  |
| 96  | 0.791 | 0.9468 | W  |
| 97  | 0.782 | 0.9678 | HW |
| 98  | 0.780 | 0.9734 | HW |
| 99  | 0.778 | 0.9782 | VW |
| 100 | 0.777 | 0.9816 | HW |
| 101 | 0.775 | 0.9861 | HW |

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