DEC 21 1965

LA-AC-7278 (Supplement)
CONF-651102-13

85622

RELEASED FOR ANNOUNCEMENT
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Supplement to "Studies on the Equilibrium Temperature for the Plutonium Alpha-Beta Transformation", by J. W. Anderson, University of California, Los Alamos Scientific Laboratory*

The cited paper presents data which establishes the equilibrium temperature for the alpha-beta transformation of Pu at $117.0 \pm 0.5^\circ\text{C}$. This was observed by following the electrical resistance change in thin, high purity, mixed-phase specimens as they were thermally cycled near the transformation temperature. As expected the time required for small phase changes near the equilibrium temperature was long. Data regarding the mechanisms of the alpha-beta and beta-alpha transformations or rates as a function of temperature were not presented.

The purpose of this supplement is to present an additional chart on this transformation and also discuss some effects of purity, thermal history and shape on the behavior of Pu specimens.

Figure 1 shows the resistance change vs. time for a 0.006 in. thick, 0.25 in. wide, 8.5 in. long ribbon of high purity Pu as it was cycled through the alpha-beta transformation in a Silicone oil bath. In the chart 100% resistance change represents complete transformation to beta-phase Pu. The specimen was heated and held at temperature for about 15 min. in 5°C increments in the alpha range. The resistance stabilized and remained constant during these hold periods up through the 115.5°C increment. As the temperature was then raised to and held at 120°C (3°C above the transformation temperature), the resistance changed rapidly. Complete transformation to the beta phase was assured by heating to 140°C . The specimen was then cooled and held at various temperatures. The resistance at 120.5°C during the cooling cycle was used to represent 100% beta phase at the transformation temperature.

*Work performed under the auspices of the United States Atomic Energy Commission.

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On cooling there was no indication of beta-alpha transformation during the 45 min. hold at 115°C (2°C below the transformation temperature). After 150 min. at 110°C the beginning of the beta-alpha transformation was detected. Subsequent cooling resulted in further transformation.

Following the termination of the run, the density of the specimen was determined daily for a week. After 2 d. it had stabilized at 19.61 g/cc compared to the original density of 19.73 g/cc. This decrease is attributed to some retained beta phase, and primarily to increases in the number and sizes of internal voids. These voids, incidentally, have very little effect on the resistance measurement because the cross-sectional mass along the specimen remains essentially constant.

Figure 2 is a plot of the potential change at constant current over an 8 in. long, 0.006 in. thick, 0.25 in. wide high purity specimen vs. temperature. This specimen, a duplicate of the above, was similarly heated (in 5°C increments and held at temperature for 15 min.) to 115°C and then cooled to ambient temperature by removal from the bath into the atmosphere of the glovebox. The potential change decreased linearly with increasing temperature up to a break between 65° and 70°C, and then continued linearly up to 115°C. As the specimen was cooled its potential change was a linear increase retracing the 70° to 115°C portion of the curve. The specimen density was 19.74 g/cc after the run compared to a beginning, 19.73 g/cc. On subsequent repeated heating cycles to 115°C, the specimen showed the potential change curve coinciding with that of the original cooling curve, there being no break. The density did not change.

Figure 3 shows the potential change vs. temperature for a similar 0.010 in. thick specimen heated to 115°C at 1°/min. The same break occurred during heating but at a higher temperature (100°-105°C) and the potential change during cooling followed the dotted line. On subsequent heating and cooling cycles to 115°C, the

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specimen showed the same potential change curve on heating and cooling as the original cooling curve. There was no change in density after the initial increase from 19.72 to 19.74 g/cc after the first cycle.

Cold cycles to -40°C on freshly cast and machined (0.005 to 0.010 in. thick) specimens of the same high purity material (Lot No. HOO-1900) or on the above two specimens after the heating cycles resulted in no measurable density changes. The increased density from the initial heating cycle and the absence of change after cold cycling in these high purity specimens is interpreted as additional alpha or beta phase (or both) being formed from retained higher phases. This occurred when internal stresses were relieved during heating.

Similar studies were made on less pure Pu. Figure 4 shows the potential change vs. temperature for a 0.006 in. thick, 0.25 in. wide 8 in. long Pu specimen heated in 5°C increments and held at temperature for 15 min. There is no break in the heating curve; however, the cooling curve shows a greater potential change from $\sim 105^{\circ}\text{C}$ to ambient temperature. This cycle resulted in a density increase of from 19.51 to 19.53 g/cc. Repeated cycles to 115°C resulted in smaller, but nevertheless, continued density increases.

Cold cycling of similar less pure Pu specimens (which had not been heat cycled) to -40°C resulted in a density increase of from 19.51 to 19.56 g/cc. This behavior is primarily attributed to the transformation to alpha or beta phase (or both) from higher phases which had been retained by specific impurities such as Ga or Si, present in this so-called less pure Pu.

Another effect on density is illustrated in Fig. 5 which shows the density changes of a high purity Pu specimen as it was progressively machined from a thickness of 0.170 to 0.005 in. The increase from 19.545 to 19.715 g/cc is a large significant change which occurred mainly between the 0.045 and 0.010 in. thicknesses. It resulted from the effect of the pressure of the cutting tool on the piece. Even with light cuts there is sufficient pressure to close microcracks

and change the internal stresses for a significant distance below the surface.

Thermal cycling of Pu through the alpha-beta and beta-alpha transformations always results in a decrease in density caused by an increase in voids and a small amount of retained beta phase. The density decrease varies directly with increasing purity and slower cooling rates through the beta-alpha transformation. Figure 6 shows the decreases in densities of two 1.375 in. diameter, 0.125 in. thick high purity Pu discs which were cycled between ambient temperature and 160°C. One disc was slowly cooled from 160°C at 0.3°C/min. while the other was quenched. The slowly cooled piece broke after four cycles while the quenched disc was still integral after thirteen cycles.

Specimen thickness plays an important role on transformation rates under a specific test condition. This was illustrated by data published in LAMS-3092. Figure 11 in that report, reproduced here, shows the times required for the alpha-beta transformation for various thicknesses of high purity specimens after each was immersed in an oil bath at 165°C. Figure 13 from that report shows the same data for less pure specimens. These illustrate the effects of thickness and purity on the transformation rates in this particular experiment. The difference between the times for transformation of the two purities at the same thickness are plotted against thickness in Fig. 7. The maximum difference for the various percentage changes (where 100% represents complete beta phase) occur in the 0.030 to 0.070 in. thickness range. Heat flow limitations are important above thicknesses of 0.070 in. while purity is less important below thickness of 0.030 in.

In summary, studies on the alpha-beta and beta-alpha transformations as well as many other measurements of Pu properties present unusual problems. Specimen geometry, history and purity each have an influence on the specific characteristics of a particular specimen. Differences in these undoubtedly account for many of the variations in a property reported in the literature.

Purity of Pu Used in Experiments

Elements	High Purity Lot HOO-1900 Lot HOO-563	Less Pure Lot JOO-1909
Li	< 0.05 ppm	< 0.05 ppm
Be	< 0.05	< 0.05
Bi	< 1	< 1
B	< 0.5	< 0.5
Mo	< 1	< 1
Mn	< 0.5	4
Pb	< 1	< 1
Sn	< 1	2
Cu	< 1	10
F	< 2	-
Si	9	70
Cr	< 1	13
Ni	< 2	55
Al	< 2	20
La	< 1	< 1
Na	< 1	< 2
Ca	< 5	6
Mg	< 5	< 2
Co	< 1	< 1
Zn	< 10	< 10
Zr	< 10	< 10
C	30	75
Ga	0.5	240
Fe	20	40
Ta	< 5	< 35
W	71	-
Am	32	-
Pu%	100.00%	99.93%

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FIG. 2

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DENSITY VARIATION VS. THICKNESS

FOR HIGH PURITY Pu

LOT H00-1796

WALL THICKNESS DECREASED BY MACHINING

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FIG. 7

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Fig. 11. Time for changes in resistance vs. thickness of high purity plutonium heated from $\sim 30^\circ$ to 165°C by immersion in silicone oil.

Fig. 13. Time for changes in resistance vs. thickness of good purity plutonium heated from $\sim 30^{\circ}$ to 165°C by immersion in silicone oil.