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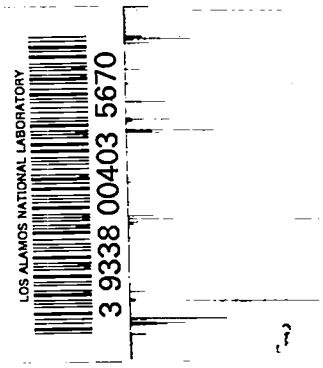
METALLOGRAPHIC PREPARATION AND OBSERVATIONS OF SOME PLUTONIUM-CARBON ALLOYS



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METALLOGRAPHIC PREPARATION AND OBSERVATIONS OF SOME PLUTONIUM-CARBON ALLOYS

by

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ABSTRACT

The metallographic preparation of plutonium-carbon alloy samples having compositions in the range 38 to 57 a/o carbon is described; and some of the optical microscopy, microhardness, volume fraction analysis, and X-ray diffractometry results thus obtained are presented.

INTRODUCTION

The poor quality and rapid deterioration of polished and etched surfaces of plutonium-carbon alloys have been obstacles to the effective optical microscopy of these alloys and have hindered metallographic confirmation of details of the plutonium-carbon phase diagram (see Fig. 1). Recently, however, a technique was developed that has routinely produced metallographic results of acceptable quality. The purpose of this report is to describe this new technique and to present some of the optical microscopy, microhardness, volume fraction analysis, and X-ray diffractometry results thus obtained.

EXPERIMENTAL AND METALLOGRAPHIC TECHNIQUES

Alloy Preparation

The specimens were prepared as arc-cast buttons by co-melting plutonium and spectroscopic grade carbon in an inert atmosphere arc furnace. The buttons, each weighing about 8 g, were inverted and remelted three times to ensure their homogeneity. Weight losses during the preparation averaged about 25 mg and were ascribed to the loss of small chips broken from the buttons during the second and third remeltings.

Mounting

The specimens for metallographic examination were mounted in the following manner. Each specimen, a sawed or cleaved section of a button,

was placed within a mold consisting of a Teflon ring on a Teflon sheet and covered with a liquid polyester plastic. After the plastic had solidified and cured overnight at room temperature, the mounted specimen was easily removed from the Teflon ring. (The use of Teflon greatly facilitates removal of the mounted specimens from the molds and allows the molds to be reused many times.) A hole was drilled through the back of the plastic mount to allow electrical contact to be made between the specimen and a metal probe used during the subsequent electrolytic polishing and etching.

Grinding

The mounted specimens were manually ground on silicon carbide papers (100, 180, 240, 320, 400, and 600 grit sizes). The essential difference between this step and the usual grinding procedure is that the specimens were ground dry, which invariably resulted in a bright, shiny specimen surface. The grinding was done in glove boxes containing "normal" laboratory air of approximately 50% relative humidity. The samples were not mechanically ground or polished beyond the 600 grit stage.

Electropolishing and Electroetching

After being ground, the specimens were simultaneously electrolytically polished and etched at 20 V (DC) for 1 min in the following solution: 133 ml acetic acid, 25 g CrO_3, and 7 ml of water. Though the resulting surfaces were slightly undulant, they were smooth and sufficiently nonreactive in air to be subjected to metallographic examination over periods of several hours, except for the specimens containing Pu_2C_3. The alloys containing Pu_2C_3, in particular the 57 a/o carbon composition, were more reactive; but use of the above technique allowed them to be examined for as long as about an hour at one time. If more than an hour was required to finish the examination, a 10 sec re-etch at 20 V was necessary.

METALLOGRAPHIC RESULTS AND OBSERVATIONS

Hardness Measurements

Microhardness measurements were made with a Leitz microhardness tester in the open laboratory; the spread of contamination was avoided by using disposable platens to support the specimens. The load was standardized at 25 g. Since both PuC and Pu_2C_3 are hard, the indentations were small, which resulted in relatively large errors in the measurements and contributed in part to the large scatter among the hardness numbers obtained for any single phase in each specimen.

PuC. The microhardness results for PuC are listed in Table I. The few unusually low values were probably due to the presence of a very fine precipitate of zeta, indicated by striations, in the PuC. Since zeta and PuC with striations are always soft and since striated PuC is found only in alloys which if in equilibrium would contain zeta, it seems likely that the striated PuC contains an extremely fine precipitate of zeta (see Fig. 2). Assuming that this explanation is correct, the data then indicate that the hardness of PuC increases with increasing carbon content, very roughly from about 500 to 900 DPH. Since the range of values obtained from any one specimen was large, the hardness may be orientation dependent.

Zeta. Many hardness measurements were made on zeta blades, and all values fell between 108 and 150 DPH. However, since each indentation occupied the entire width of a zeta blade it is probable that the harder matrix, alpha plutonium or PuC, helped to support the indenter and that the true hardness of zeta is less than 100 DPH.

Volume Fraction Analysis

Volume fraction measurements were made by means of the systematic point counting method on entire specimen sections. These measurements, corrected for the densities of the phases being considered, were plotted

vs alloy composition in w/o. Extrapolations from the plot of the measurements of the specimens heat treated at 555 and 610°C indicated a small solubility of plutonium in Pu_2C_3 (see Table II). However, the specimens heat treated at 855°C contained considerable PuC precipitate within the Pu_2C_3 dendrites (Fig. 3). This precipitation of PuC in Pu_2C_3 at 855°C indicates that the small equilibrium solubility of plutonium in Pu_2C_3 (~ 1 a/o) may occur at a somewhat higher temperature, perhaps at the peritectic temperature. No precipitated PuC was observed in the 555 and 610°C specimens, but this may be due to insufficient time at the heat treating temperatures.

X-ray Diffractometry

The phases identified by diffractometry were the same as those identified by optical microscopy (see below). Three good traces of zeta were obtained which showed several lines not reported by Mulford et al.[1] Data from these traces are listed in Table III.

Optical Microscopy

The results of optical microscopy are summarized in Table IV, and a few typical microstructures are illustrated in Figs. 4 through 9.

Zeta Peritectoid Temperature. The zeta peritectoid temperature reported as 575°C by Mulford et al.[1] and as 558°C by Rosen et al.[2] was redetermined by means of optical microscopy, diffractometry, and microhardness measurements. A large 25 g homogeneous alloy button containing 38 a/o carbon was examined in the cast condition. It was then sectioned to form several specimens, all of which were heat treated at 557°C for 2,112 hr. The specimens were quenched and found to consist of zeta plus a small amount of alpha plutonium. Three specimens were then annealed, one each at 565, 577, and 585 ± 2°C for 43 hr and quenched. Hardness and X-ray diffractometry results indicated that those annealed at 565 and 577°C still consisted of zeta plus a small amount of alpha. The specimen annealed at 585°C, however, consisted only of PuC and alpha. It follows that the zeta peritectoid

lies between 577 and 585°C.

SUMMARY

The use of dry grinding in preference to wet grinding and an electrolytic polish-etch instead of mechanical polishing has simplified the metallographic preparation of PuC alloys and has facilitated their examination. Data obtained from alloys thus prepared indicate that the hardness of PuC increases from about 500 to 900 DPH with increasing carbon content, that the true hardness of zeta is probably less than 100 DPH, and that the hardness of Pu_2C_3 is approximately 1100 DPH.

Volume fraction analysis data indicate that Pu_2C_3 will dissolve approximately 1 a/o plutonium at some temperature above 855°C. The maximum carbon content of the PuC phase appears to be 48 ± 0.5 a/o carbon according to the volume fraction measurements and slightly less than 48 a/o carbon according to optical metallography. The minimum carbon content of PuC, according to optical metallography, appears to lie between 42 and 43 a/o carbon at 610°C and between 43 and 44 a/o carbon at 550°C.

A few lines not previously reported were detected on several good diffractometer traces of zeta.

The zeta peritectoid temperature, as determined metallographically with equilibrated and heat treated specimens, appears to lie between 577 and 585°C.

REFERENCES

1. R. N. R. Mulford, F. H. Ellinger, G. S. Hendrix, and E. D. Albrecht, "The Plutonium-Carbon System," Plutonium 1960, Cleaver-Hume Press Limited, London (1960) pp. 301-311.
2. S. Rosen, M. V. Nevitt, and A. W. Mitchell "Metallographic and X-ray Observations of Pu-C Alloys," J. Nucl. Mat., 10 [2] 90-98 (1963).

Table I

Microhardness Values[a] in DPHN (25 g load) for Some Plutonium-Carbon Alloys

		PuC			Pu_2C_3		
			Heat treated			Heat treated	
a/o C	Zeta	As Cast	555°C	610°C	As Cast	555°C	610°C
38	108-150	363[b]		208[b]			
39	110-145	331[b]		245[b]			
40		741	491	689			
41		688	543	576			
42	113-150	273[b]		371[b]			
43		724	735	241[b]			
44		825	424	500			
45		835	744	789			
46		839		775			
48		919	931	834			
49		1004	714	671	1018	1161	
51		838	885	719	1051	1127	956
57			840		1029	1204	1112

[a] The reported values are averages of at least five readings, which varied as much as ± 50 DPHN in the same specimen.

[b] These unusually low values can probably be attributed to the presence of a fine precipitate of zeta.

Table II

Two-Phase Region Compositional Limits Obtained by

Extrapolating Volume Fraction Analysis Data

Temperature, °C		ε-Pu + PuC, a/o C	PuC ± Pu_2C_3, a/o C
555			47.8 ± 0.7 to 59.6 ± 0.4
610			48.2 ± 0.4 to 58.8 ± 0.4
	ε-Pu	assumed to contain 0	
	PuC	43.5 to 45.09[a]	
855			47.5 ± 0.7 to 58.6 ± 0.3[b]

[a] The reason for the range of values is not known.

[b] This compositional limit suggests some solubility of plutonium in Pu_2C_3. However, the value for the upper limit (58.6 a/o carbon) is known to be high because the method used did not allow separate measurement of a small amount of PuC that had precipitated within the measured volumes of Pu_2C_3 (see Fig. 3).

Table III

Diffractometer Results in d(Å)

Zeta Powder Pattern of Mulford et al.[1]	38 a/o C	39 a/o C	43 a/o C
2.915			
PuC	2.882	2.880	---
2.825	2.879	2.879	2.879
2.693	2.712	2.706	2.708
2.591	2.683	2.679	2.673
PuC	---	2.471	2.473
2.447	2.455	2.454	2.453
---	2.193	---	---
1.881	1.890	1.890	---
PuC	1.748	1.750	1.751
1.737	---	---	---
---	1.729	1.226	---
1.704	1.711	1.710	---
---	1.581	1.581	---
1.562	---	1.565	---
---	1.515	1.514	---
PuC	1.493	1.492	1.493
1.482	1.483	1.482	---
---	---	1.469	---
1.438	1.441	1.441	---
PuC	1.428	1.429	---
1.421	---	---	---
1.358	---	1.358	---
1.228	1.229	1.229	---
---	1.207	---	---
1.162	---	---	1.163
1.144	---	1.144	---
PuC	---	---	1.138
1.130	---	---	---
1.113	---	1.114	---
---	1.111	1.111	---
1.099	---	---	---
1.075	---	---	---

Table IV

Optical Microscopy Results for the As-Cast and Heat-Treated Plutonium-Carbon Alloys

Composition, a/o C	As Cast	H.T. at 555°C	H.T. at 610°C
38	α-Pu + PuC[a]	α-Pu + Zeta	α-Pu + PuC[a]
39	α-Pu + PuC[a]	α-Pu + Zeta	α-Pu + PuC[a]
40	α-Pu + PuC	α-Pu + Zeta + PuC	α-Pu + PuC
41	α-Pu + PuC	α-Pu + Zeta + PuC	α-Pu + PuC
42	α-Pu + PuC[a]	α-Pu + Zeta + PuC	α-Pu + PuC[a]
43	α-Pu + PuC	Zeta + PuC	PuC[a]
44	α-Pu + PuC	PuC	PuC
45	α-Pu + PuC	PuC	PuC
46	PuC + Pu_2C_3	b	PuC
48	PuC + Pu_2C_3	PuC + Pu_2C_3	PuC + Pu_2C_3
49	PuC + Pu_2C_3	PuC + Pu_2C_3	PuC + Pu_2C_3
51	PuC + Pu_2C_3	PuC + Pu_2C_3	PuC + Pu_2C_3
57	PuC + Pu_2C_3	PuC + Pu_2C_3	PuC + Pu_2C_3

[a] Also contained zeta precipitate.

[b] No result because capsule failed and specimen oxidized.

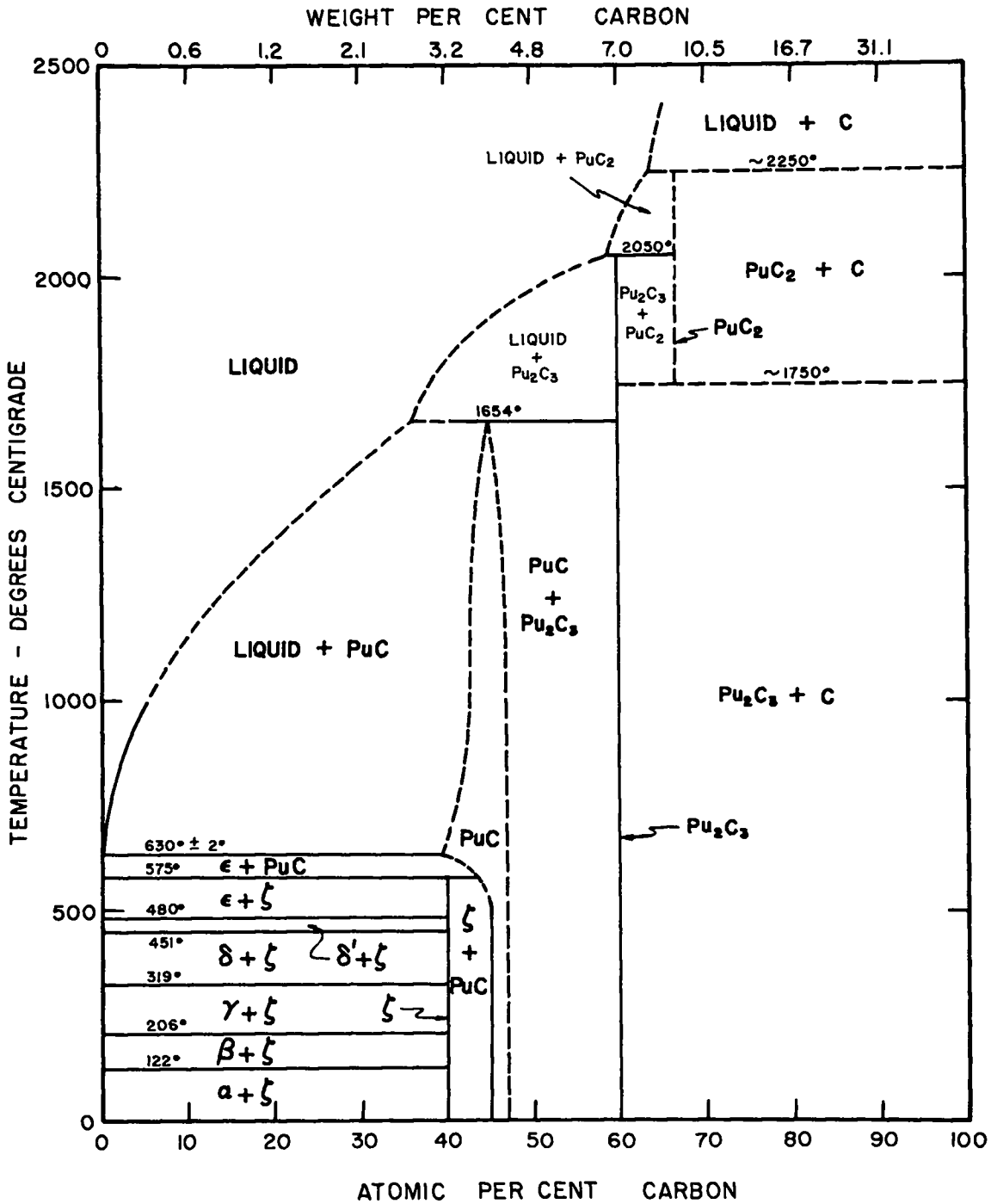

Figure 1. Plutonium-carbon phase diagram of Mulford et al.[1]

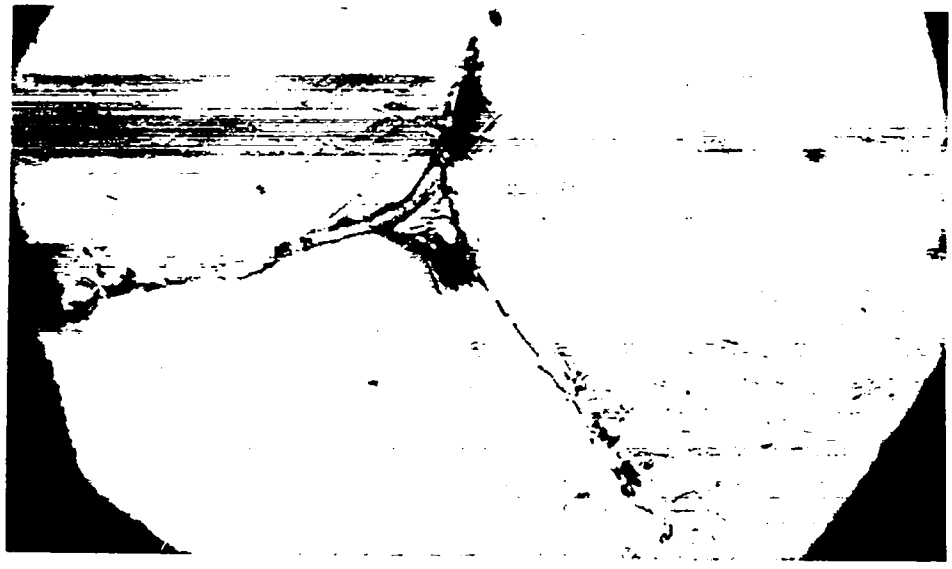

Figure 2. Plutonium-42 a/o carbon alloy as cast. 1000X. Note the fine Widmanstätten precipitate of zeta in the PuC matrix.

Figure 3. Plutonium-57 a/o carbon alloy, heat treated at 855°C for 624 hr. 500X. Note the precipitate of PuC in the Pu_2C_3 dendrites.

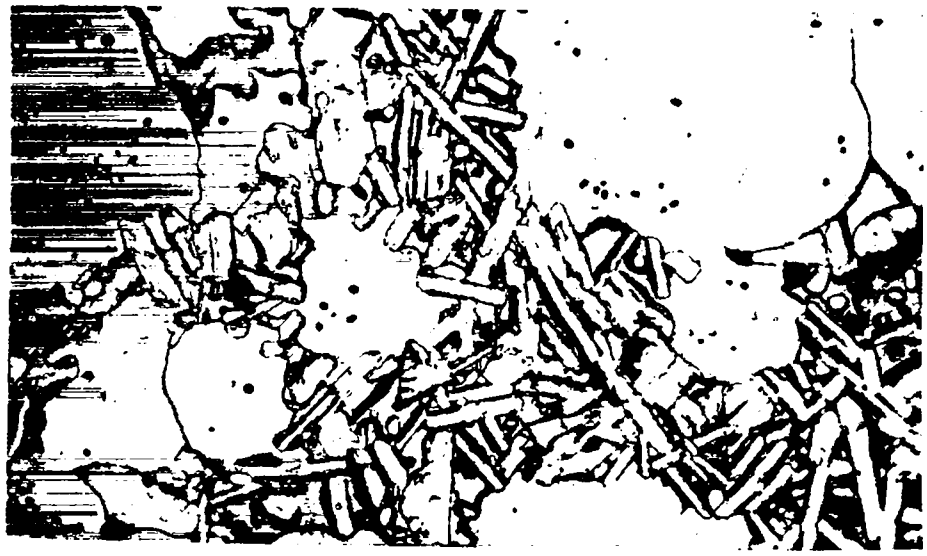

Figure 4. Plutonium-40 a/o carbon alloy, heat treated at 555°C for 840 hr. 500X. PuC grains with zeta blades growing into the alpha plutonium (dark intergramular material).

Figure 5. Plutonium-39 a/o carbon alloy, heat treated at 555°C for 864 hr. 500 X. Zeta blades in a matrix of alpha plutonium.

Figure 6. Plutonium-39 a/o carbon alloy, heat treated at 610°C for 817 hr. 500X. PuC grains with striations from zeta precipitate in an alpha plutonium matrix (dark intergranular material).

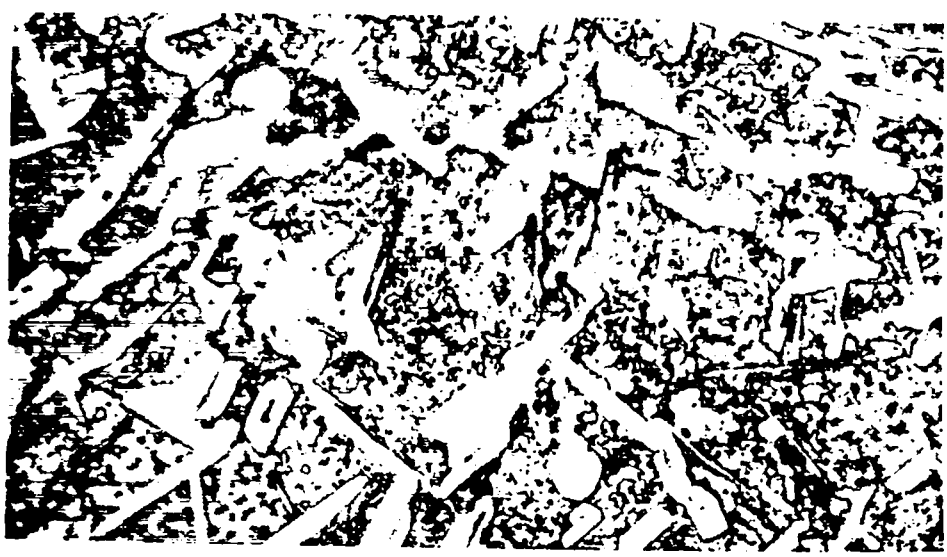

Figure 7. Plutonium-43 a/o carbon alloy, heat treated at 555°C for 864 hr. 500X. Zeta grains in a PuC matrix.

Figure 8. Plutonium-48 a/o carbon alloy, heat treated at 555°C for 818 hr. 500X. Note the residual Pu_2C_3 left after the heat treatment.

Figure 9. Plutonium-57 a/o carbon alloy, heat treated at 555°C for 794 hr. 500X. Pu_2C_3 dendrites in a PuC matrix.