BNWL-1545
UC-25

X-RAY COMPARISON
OF BETA PLUTONIUM GRAIN SIZE

R. P. Allen
H. W. Arrowsmith
R. D. Nelson

January 1971

AEC RESEARCH &
DEVELOPMENT REPORT

BNWL-1545

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

PACIFIC NORTHWEST LABORATORY

RICHLAND, WASHINGTON

operated by

BATTELLE MEMORIAL INSTITUTE

for the

UNITED STATES ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(45-1)-1830

3 3679 00061 7748

BNWL-1545

UC-25, Metals, Ceramics,
and Materials

X-RAY COMPARISON OF BETA PLUTONIUM GRAIN SIZE

By

R. P. Allen, H. W. Arrowsmith and R. D. Nelson

Metallurgy Development Department
Chemistry and Metallurgy Division

January 1971

BATTELLE MEMORIAL INSTITUTE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

Printed in the United States of America
Available from
National Technical Information Service
National Bureau of Standards,
U.S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

X-RAY COMPARISON OF BETA PLUTONIUM GRAIN SIZE

R. P. Allen, H. W. Arrowsmith and R. D. Nelson

ABSTRACT

The relative grain size of alpha plutonium, beta plutonium formed from the alpha phase (β_α), beta plutonium formed from the gamma phase (β_γ), and gamma plutonium was determined using an elevated-temperature X-ray metallographic technique. Fine-grained alpha that was transformed $\alpha \rightarrow \beta \rightarrow \gamma \rightarrow \beta \rightarrow \alpha$ at 3 °C/min gave fine-grained β_α , coarse-grained gamma, and coarse-grained β_γ . The alpha formed from the coarse-grained β_γ and the subsequent β_α were both coarsened by the gamma phase heat treatment. Conversely, $\alpha \rightleftharpoons \beta$ transformation cycling refined the grain size of the alpha and the β_α . A specimen of coarse-grained alpha (300 to 500 μ average grain diameter) was slowly transformed $\alpha \rightarrow \beta$ at 5 °C/hr to maximize the β_α grain size and then rapidly transformed $\beta \rightarrow \gamma \rightarrow \beta$ at 20 °C/min to minimize the β_γ grain size. The resultant β_α was again much finer grained than the β_γ , confirming the existence of a characteristic difference in the average grain size of β_α and β_γ .

CONTENTS

LIST OF FIGURES	vii
INTRODUCTION	1
EXPERIMENTAL PROCEDURE	2
RESULTS AND DISCUSSION	3
CONCLUSIONS	6
ACKNOWLEDGEMENT	6
REFERENCES	11
DISTRIBUTION	12

LIST OF FIGURES

1	α -- 25 °C	7
2	β_{α} -- 140 °C	7
3	γ -- 230 °C	7
4	Mixture of γ and β_{γ}	7
5	β_{γ} -- 120 °C	8
6	α -- 25 °C	8
7	Mixture of α and β_{α}	8
8	β_{α} --- 180 °C	8
9	γ -- 220 °C	9
10	β_{γ} -- 110 °C	9
11	α -- 95 °C	9
12	β_{α} -- 128 °C	9
13	α -- 25 °C	10
14	β_{α} -- 155 °C	10
15	α -- 25 °C	10
16	β_{α} -- 160 °C	10

X-RAY COMPARISON OF BETA PLUTONIUM GRAIN SIZE

R. P. Allen, H. W. Arrowsmith and R. D. Nelson

INTRODUCTION

An elevated-temperature X-ray metallographic technique was used to determine the relative grain size of beta plutonium formed from the lower temperature alpha phase, and beta plutonium formed from the higher temperature gamma phase. Beta formed from alpha is termed β_α and beta formed from gamma is termed β_γ . The $\alpha \rightleftharpoons \beta$ transition occurs at about 115 °C and the $\beta \rightleftharpoons \gamma$ transition occurs at about 190 °C. Alpha plutonium has a monoclinic crystal structure with 16 atoms per unit cell, beta plutonium is body-centered monoclinic with 34 atoms per unit cell, and gamma plutonium is face-centered orthorhombic with 8 atoms per unit cell.⁽¹⁾

Nelson, Bowman, and Bierlein⁽²⁾ studied the deformation and phase transformation behavior of high-purity beta plutonium and found marked differences in both the mechanical properties and the $\beta \rightarrow \alpha$ transformation behavior of β_α and β_γ . Beta formed from gamma, for example, was consistently stronger and far less ductile than beta formed from alpha. Similarly, the $\beta_\gamma \rightarrow \alpha$ transformation occurred at a much faster rate than the corresponding $\beta_\alpha \rightarrow \alpha$ transformation at all test temperatures. Dahlgren,⁽³⁾ in a subsequent study of the plasticity of beta plutonium, found that β_α exhibited the superplastic behavior and the high values of the strain-rate-hardening exponent characteristic of a material with a fine-grained microstructure. However, β_γ exhibited significantly lower values of the strain-rate-hardening exponent, and the type of brittle behavior characteristic of a material with a coarse-grained microstructure.

This gross difference in the plasticity of beta plutonium thus constitutes indirect evidence of a difference in the characteristic grain size of β_α and β_γ .

The $\beta \rightarrow \alpha$ transformation behavior of beta plutonium can also be rationalized in terms of a difference in beta grain size. The $\beta \rightarrow \alpha$ transformation rate should be greater for coarse-grained beta if the amount of alpha formed per nucleation event is a function of grain size. This type of grain size dependency is observed for the martensitic $\beta \rightarrow \alpha$ transformation in dilute U-Cr alloys.⁽⁴⁾ Unfortunately, all attempts to verify the apparent difference in the characteristic grain size of β_α and β_γ using conventional elevated temperature metallographic techniques have been unsuccessful. Because of the disruptive effect of the 9% volume change associated with the $\alpha \rightleftharpoons \beta$ transformation, it is necessary to polish, etch, and examine specimens at beta phase temperatures. Arrowsmith⁽⁵⁾ developed a successful technique for polishing plutonium in the beta phase, and some beta grain structure has been developed by the at-temperature deformation of polished beta surfaces.⁽⁶⁾ However, all efforts to electrochemically etch the polished beta have failed because of the extreme chemical reactivity of plutonium at the required temperatures. An elevated-temperature X-ray metallographic approach was used to circumvent these difficulties and provide at least a qualitative comparison of the relative grain size of β_α and β_γ .

EXPERIMENTAL PROCEDURE

At-temperature X-ray diffraction photographs of alpha, beta, and gamma plutonium were obtained using a standard forward-reflection flat-film camera technique. The camera system was designed so that successive diffraction photographs

could be made without changing the position of the specimen or altering any of the test parameters other than specimen temperature. All of the alpha, beta, and gamma patterns for a particular specimen were thus taken from the same location on the specimen to permit a direct comparison of the relative grain size for these phases. Exposures were made using both filtered and unfiltered molybdenum radiation with the diffraction surface inclined at an angle of 11 degrees to the incident X-ray beam. The diffraction specimens were prepared from high-purity, electrorefined plutonium containing less than 300 ppm total impurities. The specimens were electropolished and positioned in a straight-through type vacuum furnace sealed with 0.75 mm Teflon windows. The furnace could be heated or cooled at a maximum rate of 20 °C/min and had a maximum temperature capability of 250 °C. A molecular-sieve cold trap was used with the furnace to eliminate water vapor and retard oxidation of the specimens. Details of specimen preparation and phase transformation conditions will be presented in the next section.

RESULTS AND DISCUSSION

The diffraction photographs for two successive $\alpha \rightarrow \beta \rightarrow \gamma \rightarrow \beta \rightarrow \alpha$ cycles are shown in Figures 1 through 10. The diffuse central reflections common to all of the patterns are from the Teflon vacuum seal and the stainless steel furnace liner. The remainder of the diffraction lines are those of the plutonium. Only relative grain sizes can be estimated from the appearance of the patterns. The assignment of absolute β grain size values would require a set of β grain size standards. However, for conventional materials and experimental conditions the spotty-to-continuous transition in the appearance of a diffraction pattern occurs around the 1 to 10 μ grain size range.⁽⁷⁾

Tests using alpha plutonium gave an average grain size of 10 to 15 μ for the transition region. It will therefore be assumed that distinctly spotty β patterns represent an average grain size substantially greater than these transition values and that β patterns with smooth, continuous diffraction lines represent an average grain size substantially smaller than the maximum transition values.

The specimen used for the $\alpha \rightarrow \beta \rightarrow \gamma \rightarrow \beta \rightarrow \alpha$ cycles was cooled $\gamma \rightarrow \beta \rightarrow \alpha$ at 20 °C/min to give an initially fine-grained alpha structure (Figure 1). Transforming this fine-grained alpha to beta at a heating rate of 3 °C/min gave an equally fine-grained β_{α} structure (Figure 2). The same 3 °C/min heating and cooling rate was used for the subsequent transformation cycles. The gamma formed from the fine-grained β_{α} was coarse grained as shown in Figure 3. The diffraction pattern shown in Figure 4 was made during the $\gamma \rightarrow \beta_{\gamma}$ transformation. The gamma lines are still present and additional spots corresponding to the beta phase have appeared. The pattern obtained at the conclusion of the $\gamma \rightarrow \beta_{\gamma}$ transformation (Figure 5) shows that the β_{γ} formed from the coarse-grained gamma is also coarse-grained. Similarly, the alpha formed from the coarse-grained β_{γ} (Figure 6) has a coarser grain size than the initial alpha. Both the alpha and the beta structures were therefore coarsened by the gamma phase heat treatment.

The results for the second $\alpha \rightarrow \beta \rightarrow \gamma \rightarrow \beta$ cycle are given in Figures 7 through 10. The $\alpha \rightarrow \beta_{\alpha}$ transformation pattern (Figure 7) and the final β_{α} pattern (Figure 8) show that the β_{α} formed from the coarser-grained alpha is generally fine grained but also contains some discrete, larger grains comparable in size to those of the prior β_{γ} . The gamma pattern (Figure 9) is similar to that for the first gamma cycle

(Figure 3). While the β_γ for the second cycle (Figure 10) has a slightly finer grain size than the initial β_γ (Figure 5) it is still definitely coarse grained.

These X-ray results show that the grain size of β_γ is larger than the grain size of β_α for the particular $\alpha \rightarrow \beta$ and $\gamma \rightarrow \beta$ transformation conditions employed in the study. A third $\alpha \rightarrow \beta \rightarrow \gamma \rightarrow \beta$ cycling test was therefore conducted using a special specimen with large alpha grains (300 to 500 μ) to determine if, by altering the transformation conditions, β_α and β_γ with a comparable grain size could be produced. The specimen* was prepared by cooling electrorefined plutonium $\beta \rightarrow \alpha$ at 0.25 °C/hr while under a compressive stress of 50,000 psi. The specimen was then slowly transformed $\alpha \rightarrow \beta$ at 5 °C/hr to maximize the β_α grain size. Figures 11 and 12 show that appreciable grain refinement occurred to give β_α with an average grain size still substantially finer than the β_γ of Figures 5 and 10. The specimen was then rapidly transformed $\beta \rightarrow \gamma \rightarrow \beta$ at a heating and cooling rate of 20 °C/min to minimize the β_γ grain size. The photograph of the resulting β_γ pattern was not suitable for publication but the pattern represented an average grain size only slightly finer than that shown in Figure 10. Although both β_α and β_γ show some variation in grain size with either transformation conditions or the grain size of the parent phase, these results clearly demonstrate that the grain size of β_α is characteristically finer than the grain size of β_γ .

An $\alpha \rightarrow \beta \rightarrow \alpha \rightarrow \beta$ cycling test was also conducted to determine if the grain size of β_α is reduced by successive $\alpha \rightleftharpoons \beta$ transformations. A decrease in the grain size of alpha with $\alpha \rightleftharpoons \beta$ cycling has been observed metallographically.⁽⁸⁾ This

---

* Furnished by M. D. Merz, Metallurgy Development Department

refinement of the alpha grain size should result in a corresponding decrease in the grain size of the β_α . Grain refinement of the β_α would be consistent with the larger value of the strain-rate-hardening exponent noted by Dahlgren⁽³⁾ for beta plutonium that had been $\alpha \rightarrow \beta \rightarrow \alpha \rightarrow \beta$ cycled. Grain refinement of the β_α would also explain the progressive increase in the incubation time for the $\beta \rightarrow \alpha$ transformation reported by Allen and Nelson⁽⁹⁾ for specimens cycled $\alpha \rightleftharpoons \beta$ after an initial gamma phase heat treatment. Figures 13 through 16 show the diffraction patterns for the $\alpha \rightarrow \beta \rightarrow \alpha \rightarrow \beta$ cycling test. The initial alpha was formed by cooling the specimen from the beta phase at 1 °C/hr. The specimen was heated $\alpha \rightarrow \beta$ at 0.3 °C/min and cooled $\beta \rightarrow \alpha$ at 1 °C/min. A comparison of the relative grain size of the alpha (Figure 13 and 15) and of the β_α (Figure 14 and 16) shows that appreciable grain refinement occurred for both the β_α and the alpha as a result of the $\alpha \rightleftharpoons \beta$ transformation cycling.

CONCLUSIONS

1. Beta plutonium formed from coarse-grained gamma plutonium has a characteristically larger average grain size than beta plutonium formed from either coarse-grained or fine-grained alpha plutonium.
2. The average grain size of alpha and β_α plutonium is increased by prior gamma phase heat treatment and is reduced by subsequent $\alpha \rightleftharpoons \beta$ transformation cycling.

ACKNOWLEDGEMENT

The authors wish to thank H. E. Kissinger for helpful suggestions and comments concerning both the experimental portion of this study and the interpretation of the diffraction photographs.

FIGURE 1. α -- 25 °C

FIGURE 2. β_{α} -- 140 °C

FIGURE 3. γ -- 230 °C

FIGURE 4. Mixture of γ and β_{γ}

FIGURE 5. β_{γ} -- 120 °C

FIGURE 6. α -- 25 °C

FIGURE 7. Mixture of α and β_{α}

FIGURE 8. β_{α} -- 180 °C

FIGURE 9. γ -- 220 °C

FIGURE 10. β_{γ} -- 110 °C

FIGURE 11. α -- 95 °C

FIGURE 12. β_{α} -- 128 °C

FIGURE 13. α -- 25 °C

FIGURE 14. β_{α} -- 155 °C

FIGURE 15. α -- 25 °C

FIGURE 16. β_{α} -- 160 °C

REFERENCES

1. W. N. Miner and F. W. Schonfeld. "Physical Properties," Plutonium Handbook 1, edited by O. J. Wick. Gordon and Breach Science Publishers, New York, 1967. pp. 33-57.
2. R. D. Nelson, F. E. Bowman, and T. K. Bierlein. "Transformation and Deformation in Plutonium," Plutonium 1965, edited by A. E. Kay and M. B. Waldron. Chapman and Hall, London, 1967. pp. 1-17.
3. S. D. Dahlgren. "Superplasticity of Unalloyed Beta Plutonium," Trans. Met. Soc. AIME, vol. 242, pp. 126-132. January 1968.
4. R. D. Townsend and J. Burke. "The Effects of Thermal History on the Kinetics of the $\beta \rightarrow \alpha$ Phase Change in Uranium and Uranium-Chromium Alloys," J. Nucl. Mater., vol. 17, pp. 215-226. 1965.
5. H. W. Arrowsmith. "Elevated Temperature Metallography of Beta Plutonium," to be published in Proceedings of the 23 Annual AEC Metallographic Meeting, 1969.
6. R. P. Allen, H. W. Arrowsmith, and R. D. Nelson. "Grain Size Dependency of Beta-Plutonium Plasticity," Met. Trans., vol. 2. 1971.
7. A. Taylor. X-Ray Metallography. John Wiley and Sons, Inc., New York, 1961. pp. 663-671.
8. M. Rosen, L. T. Lloyd and R. G. Petersen. "The $\beta \rightarrow \alpha$ Phase Transformation in High Purity Plutonium," Plutonium 1965, edited by A. E. Kay and M. B. Waldron. Chapman and Hall, London, 1967. pp. 18-38.
9. R. P. Allen and R. D. Nelson. "Beta Grain Size and the $\beta \rightarrow \alpha$ Transformation in Unalloyed Plutonium," Plutonium 1970 and Other Actinides, edited by W. N. Miner. AIME, New York, 1970. Part II, pp. 991-1000.

DISTRIBUTIONNo. of
CopiesOFFSITE

1	<u>AEC Chicago Patent Group</u> G. H. Lee
210	AEC Division of Technical Information Extension
1	<u>AEC Library, Washington</u> Lt. Col. H. W. Parlett Div. Military Application
6	<u>Dow Chemical Company (AEC)</u> R. D. Forest (3) J. F. Willging L. W. Pribila F. J. Fraikor
4	<u>Lawrence Radiation Laboratory, Livermore (AEC)</u> J. O. Jepson W. J. Ramsey J. L. Robbins (2)
3	<u>Los Alamos Scientific Laboratory (AEC)</u> R. D. Baker W. J. Maraman F. W. Schonfeld

ONSITE

1	<u>AEC Chicago Patent Group</u> R. K. Sharp (Richland)
1	<u>AEC Richland Operations Office</u> M. R. Schneller/J. L. Rhoades
2	<u>RDT Assistant Director for Pacific Northwest Programs</u>

No. of
Copies

74

Battelle-Northwest

R. P. Allen (25)
H. W. Arrowsmith
V. C. Asmund
R. A. Busch
S. D. Dahlgren
D. R. de Halas
H. R. Gardner
J. W. Patten
W. S. Hartnett
D. D. Hays
Y. B. Katayama
R. R. King, Jr.
M. D. Merz
E. D. McClanahan
R. W. Moss
R. D. Nelson (25)
R. W. Stewart
J. M. Taylor
O. J. Wick
Technical Information (5)
Technical Publications (2)