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Quantitative Metallography of Plutonium Using Point-Count Techniques

by
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ROCKY FLATS DIVISION

GOLDEN, COLORADO

U.S. ATOMIC ENERGY COMMISSION CONTRACT AT (29-1)-1106

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RFP-331

UC-25 METALS, CERAMICS,
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THE DOW CHEMICAL COMPANY
ROCKY FLATS DIVISION
GOLDEN, COLORADO

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QUANTITATIVE METALLOGRAPHY OF PLUTONIUM
USING POINT-COUNT TECHNIQUES

by

R. L. Greeson

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ABSTRACT

A point-counter was designed and adapted for use on a Bausch and Lomb Balphot Metallograph. The results of quantitative metallography performed on materials with two or more phases are given and some of the results are compared with x-ray analysis.

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INTRODUCTION

A point-count system using a point-grid which is generated by means of spring-clips riding on notched wheels was designed and adapted for use on a Bausch and Lomb Balphot Metallograph (see Figures 1 and 2). This point-count device was patterned after one described by F. Chayes⁽¹⁾.

SUMMARY AND CONCLUSIONS

Point-count analysis methods can be summarized as follows:

Point-Count Analysis of Pu₆Fe Intermetallics

Point-count analysis methods on Pu₆Fe inclusions are reproducible. A statistical method of comparison has shown that the means of two or more populations, as determined by several operators are not different assuming that they have unequal standard deviations.

A comparison of point-count results to analytical results is very good.

Point-Count Analysis of PuC Cermets

Point-count results and x-ray results appear to be in agreement although no direct comparison can be made at this time. We are unable, at this time, to determine the density of PuC by x-ray techniques.

Once particles have been identified, point-count results can show any volume percent change that might take place upon subsequent heat treatments.

Point-Count Analysis of Varying Amounts of Alpha Plutonium In Delta Plutonium

Percent alpha as determined by point-count techniques is in agreement with x-ray analysis when the alpha fields are equiaxed.

⁽¹⁾ F. Chayes, "A Simple Point-Counter for Thin-Section Analysis", American Mineralogist 34, 1-11 (1949).

Point-count results may be more reliable than x-ray results because orientation and segregation effects are observable and can be taken into account. In addition, the entire surface of a sample can be observed metallographically while only a small area is seen by x-ray.

Alpha forms as stringers in chill-cast delta plutonium which has been rolled to more than 30% reduction. Hence, different surfaces of the same sample may show different amounts of alpha. In thin samples, such as these, edge views can be much more easily point-counted than analyzed by x-ray techniques.

It has been learned that the amount of alpha plutonium seen on the surface of samples of delta plutonium is not representative of the alpha plutonium present in the bulk sample, but is a surface effect approximately two microns thick. This surface effect can be attributed to mechanical work during sample preparation.

EXPERIMENTAL

The test samples used in this study were routine samples, for the most part, submitted by members of the Research and Development group. Metallographic sampling was done by members of the optical and x-ray group of the Research and Development facility.

Apparatus

The metallographic point-count analyses were made on a Bausch and Lomb Balphot Metallograph which had been adapted with a point-counter. Figure 1 is a photograph of the stage and Figure 2 is a schematic of the stage. The lineal distance per grid intersection is 0.5 mm in both directions. An area, 5/8 inch square, can be covered by approximately 2000 grid intersections. If additional points are required due to sample geometry, the sample can be rotated through 360°. This changes the grid intersection points.

Method

The method used in determining the volume percentages present is based upon the relation:

$$V/o = \frac{(\text{total counts of A})}{(\text{total counts of A}) + (\text{total counts of B}) + \dots} \times 100$$

Several two-unit counters are used to obtain numbers which are proportional to the volume percents of the various phases present. The optical stage is moved discontinuously and stopped at points, determined by the ratchets in the translating mechanism as shown in Figures 1 and 2. A count is recorded on the appropriate unit whenever the hairline appears to stop on a given phase.

The statistical method of analyzing the data obtained from point-counting is based on one described by Howard and Cohen⁽²⁾. Ten to thirty groups consisting of one-hundred counts per group were used for each determination described below:

$$x_1, x_2, x_3 \dots x_n \text{ are the total (n) groups where} \quad (1)$$
$$x_i \text{ is a given group.}$$

$$\text{The mean} = \bar{x} = \frac{\sum_{i=1}^n x_i}{n} \quad (2)$$

$$\text{The variance} = V_x = \frac{\sum_{i=1}^n x_i (x_i - \bar{x})^2}{n-1} \quad (3)$$

$$\text{The standard deviation} = S_x = \sqrt{V_x} = \sqrt{\frac{\sum_{i=1}^n x_i (x_i - \bar{x})^2}{n-1}} \quad (4)$$

$$\text{The standard error} = S_{\bar{x}} = \frac{S_x}{\sqrt{n}} \quad (5)$$

(2) R. T. Howard and M. Cohen, "Quantitative Metallography by Point-Counting and Lineal Analysis", Trans. AIME 172, 413-426 (1947).

a 65% confidence limit within 1% of S_x is desired, then the following expression gives the number of determinations needed:

$$n_{65} = \pm S\bar{x} \quad (6)$$

If a 95% confidence limit within 1% of S_x is desired, then the following expression gives the number of determinations needed:

$$n_{95} = \pm 2 S\bar{x} \quad (7)$$

The following equations were used to convert volume percent Pu_6Fe inclusions to parts per million iron:

$$wt\% Pu_6Fe = \frac{(\rho_{Pu_6Fe})(V/o_{Pu_6Fe})}{(\rho_{Pu_6Fe})(V/o_{Pu_6Fe}) + (\rho_{Pu*})(V/o_{Pu})} \quad (8)$$

*The theoretical density of alpha plutonium was used.

$$wt\% Fe \text{ in } Pu_6Fe = \frac{(\text{atomic wt Fe})(1/7) \times 100}{(\text{atomic wt Fe})(1/7) + (\text{atomic wt Pu})(6/7)} = 2.92 \quad (9)$$

$$wt\% Fe \text{ in sample} = (wt\% Fe \text{ in } Pu_6Fe)(wt\% Pu_6Fe) = 2.92(wt\% Pu_6Fe) \quad (10)$$

$$g/g Fe = (wt\% Fe \text{ in sample})(10^{-2}) \quad (11)$$

$$PPM Fe = (g/g Fe)(10^6) \quad (12)$$

The following expression was used for the conversion of volume percent alpha in delta plutonium to weight percent alpha. With this value, then a direct comparison with x-ray was made.

wt% alpha

$$= \frac{(V/o \text{ alpha})(\rho \text{ of alpha}^*) \times 100}{(V/o \text{ alpha})(\rho \text{ of alpha}^*) + (V/o \text{ delta})(\text{calculated } \rho \text{ of delta}^{**})} \quad (13)$$

*The theoretical density of alpha plutonium was used.

**The density of delta plutonium was calculated from the lattice parameter (A_0) as determined by x-ray analysis.

DISCUSSION

The results obtained from all the data used in this study were analyzed using the statistical analysis described above.

The percent spread as a function of volume percent was plotted

and is shown in Figure 3. Using this graph to determine a 95% confidence limit, it is no longer necessary to take groups of points, and the time required for an operator to point-count a sample is greatly reduced.

The number of observations required for a 95% probability of $\pm 1\%$ error or less is determined by using the value $4 V_x$. The value is determined from the data obtained from each sample using equation (3). Figure 4 was plotted using the values obtained. The dotted line is a possible curve. The large scattering of points is attributable to inhomogeneity in the samples.

The following data was obtained from the studies made.

Point-Count Analysis of Pu₆Fe Intermetallics

Table I shows the point-count results obtained from point-counting and converting volume percent Pu₆Fe intermetallic to parts per million iron by using equations (8) through (12). These results are compared to analytical results.

Routine metallographic samples can be easily point-counted for volume percent Pu₆Fe along with the routine metallographic work normally performed on the sample. The approximate additional time required to make an iron analysis is about one hour.

Point-Count Analysis of PuC Cermets

Table II shows some of the results obtained from point-counting PuC cermets. Also included are the x-ray results obtained from one sample of run number 31 and two samples of run number 32.

Point-Count Analysis of Varying Amounts of Alpha Plutonium In Delta Plutonium

Table III shows the point-count results obtained to date from point-counting alpha plutonium in delta plutonium. All of the volume percents have been converted to weight percents using densities calculated from x-ray lattice parameter data and these weight percents are compared to weight percents which have been estimated from peak intensities.

In the range of 0 to 10 percent, comparison between the two methods is not good. Since x-ray does not cover the same amount

of area as point-count and if the alpha phase particles are fairly unevenly dispersed, it is conceivable that in the low ranges point-count results are much more reliable.

TABLE I

Results Obtained From Point-Counting Pu₈Fe Intermetallics

Sample No.	Analytical Results Fe (PPM)	Point-Count Results Fe (PPM)	Total Counts Taken
1	169	160 $\pm$ 16	2500
2	431	427 $\pm$ 50	1500
3	508	525 $\pm$ 53	1500
4	508	482 $\pm$ 28	2500
5	931	1225 $\pm$ 142	2000
6	1460	1607 $\pm$ 154	1500
7	3087	3361 $\pm$ 321	1500

In the range of 10 to 20 percent, very good correlation is found between the two methods. Additional samples in this range of percent alpha phase are currently being studied. First results indicate continuing good correlation between the two methods.

Above 20 percent, comparison between the two methods is not good. As an example, sample number 11 was point-counted in both the short transverse (x-ray surface) and in the long transverse directions. It was found that a much higher percentage of alpha phase was in the center of the sample than on the edges in the short transverse direction. It was also found that a large variation in percent alpha phase existed between the long and short transverse directions.

Figure 5 (B) is a typical metallographic view of alpha plutonium in a long transverse direction of a sheet of delta plutonium rolled to more than 30% reduction. Figure 5 (C) is typical of the way alpha plutonium appears metallographically in a short transverse (x-ray) direction of a rolled sheet of delta plutonium. Figure 5 (A) is typical of an "as-cast" sample.

TABLE II

Results Obtained From Point-Counting PuC Cermets

<u>Run No.</u>	<u>Sample No.</u>	<u>Point-Count V/o</u>	<u>X-Ray-Peak Area w/o</u>
31	1	74 ± 1 PuC	72 PuC
		26 ± 1 $\alpha + \delta$	14 α 14 δ
	2	76 ± 1 PuC	
		24 ± 1 $\alpha + \delta$	
	3	76 ± 1 PuC	
		24 ± 1 $\alpha + \delta$	
	4	75 ± 1 PuC	
		25 ± 1 $\alpha + \delta$	
	5	74 ± 2 PuC	
		26 ± 2 $\alpha + \delta$	
	6	74 ± 1 PuC	
		26 ± 1 $\alpha + \delta$	
	7	75 ± 1 PuC	
		25 ± 1 $\alpha + \delta$	
32	1	80 ± 1 PuC	76 PuC
		20 ± 1 $\alpha + \text{Pu}_3\text{C}_2$	19 α 5 Pu_3C_2
	2	81 ± 1 PuC	
		19 ± 1 $\alpha + \text{Pu}_3\text{C}_2$	
	3	80 ± 1 PuC	
		20 ± 1 $\alpha + \text{Pu}_3\text{C}_2$	
	4	79 ± 1 PuC	
		21 ± 1 $\alpha + \text{Pu}_3\text{C}_2$	
	5	79 ± 2 PuC	
		21 ± 2 $\alpha + \text{Pu}_3\text{C}_2$	
	6	79 ± 2 PuC	76 PuC
		21 ± 2 $\alpha + \text{Pu}_3\text{C}_2$	20 α 6 Pu_3C_2

In all cases where point-count results were compared to x-ray results, the same surface was used. If a good correlation could not be obtained between the two methods, additional surfaces of the same sample were prepared, point-counted, and analyzed. In this way it was concluded that cases do exist where, due to the stringer type formation, more alpha phase appears to be present in one direction than in another direction.

TABLE III

Results Obtained From Both Point-Count And
X-Ray Data-Delta Plutonium

Sample No.	Point-Count V/o α	Point-Count Calculated* w/o α	X-Ray Calculated** w/o α
1	7.0 $\pm$ 0.9	8.5 $\pm$ 1.1	2.1
2	10.9 $\pm$ 1.1	11.2 $\pm$ 1.1	8.8
3	11.7 $\pm$ 1.1	12.8 $\pm$ 1.2	12.1
4	12.9 $\pm$ 1.2	15.7 $\pm$ 1.4	16.3
5	15.9 $\pm$ 1.3	18.6 $\pm$ 1.5	19.2
6	17.2 $\pm$ 1.4	20.6 $\pm$ 1.6	21.4
7	18.7 $\pm$ 2.9	22.2 $\pm$ 1.6	31.2
8	22.1 $\pm$ 1.5	27.1 $\pm$ 1.9	25.2
9	23.3 $\pm$ 1.6	24.8 $\pm$ 1.9	30.1
10	25.9 $\pm$ 1.7	30.2 $\pm$ 1.9	30.4
11	27.5 $\pm$ 1.7	32.1 $\pm$ 2.0	42.0

* Point-count weight percent calculated from volume percent and calculated density.

** X-ray estimated weight percent calculated from peak intensities.

Unusually high amounts of alpha phase material were observed in these samples. We do not believe that this is representative of the bulk material, since the densities of these samples are not correspondingly high. At the present time we feel that some alpha phase material had been produced on the surface of the samples during metallographic preparation. Future work will be directed toward attempts to make surface observations which will be representative of the bulk material.

Figure 1. The Point-Count Adapter and Metallographic Stage

POINT COUNTER ASSEMBLY

Note: Shaded parts have been added to a standard metallograph stage.

Figure 2, A Schematic of the Point-Counter

Figure 3

Figure 4

(A) As Cast - Mag.250X, Longitudinal
v/o(alpha) = 14.8 ± 1.2

(B) Cast and Rolled - Mag.500X
Long Transverse - v/o (alpha)=
 22.1 ± 1.7

(C) Cast and rolled - Mag.500X
Short Transverse - v/o(alpha)=
 19.6 ± 1.6

Figure 5. Typical delta Pu after two different stages of fabrication. The black appearing structure is alpha Pu. The matrix is delta Pu. The white networks in the "as cast" sample are Pu_6Fe inclusions.