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MEASURING PERCENT ALPHA IN DELTA STABILIZED
PLUTONIUM ALLOYS BY X-RAY TECHNIQUES

by

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U.S. ATOMIC ENERGY COMMISSION
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CONTENTS

	Page No.
ABSTRACT.....	1
INTRODUCTION	1
DEVELOPMENT OF THEORY	1
DETERMINATION OF CONSTANT	2
DESCRIPTION OF METHOD.....	4
Specimen Preparation	4
Making the Measurement	4
SOURCES OF ERRORS	4
Problems of Diffraction from Plutonium	4
Errors Peculiar to the Method	5
VERIFICATION OF METHOD	6
Samples of Known Composition	6
Samples of Unknown Composition	6
DISCUSSION	7

ABSTRACT

A method for measuring the percent alpha plutonium in delta stabilized plutonium alloys is described. The method is nondestructive and utilizes an X-ray diffractometer for the quantitative measure.

INTRODUCTION

Techniques employing X-ray diffraction in the qualitative and quantitative analysis of conventional materials are well established. The purpose of this study is to develop a reliable X-ray method for measuring the percent alpha in delta stabilized plutonium alloys.

Plutonium has six allotropic modifications. In unalloyed plutonium the FCC delta phase is stable between 319°C and 450°C. By alloying with certain stabilizing elements, it is possible to render the delta phase stable or metastable at ambient temperatures.

In three binary plutonium alloy systems, namely Pu-Al,⁽¹⁾ Pu-Ce,⁽²⁾ and Pu-Ga,⁽³⁾ the delta phase is known to be stabilized to below room temperature. The phase diagrams of the three systems are comparable and show that the solutes have virtually no solubility in the alpha, beta, gamma or delta phases but are markedly soluble in the epsilon and delta phases, stabilizing the latter to room temperature.

In a number of other binary alloy systems, the delta phase can be retained to room temperature. However, in most instances it appears to be a metastable retention.

The stabilization or metastabilization of delta is often not 100 percent efficient. Frequently, one obtains a two-phase mixture consisting of delta plus alpha-phase plutonium. As would be expected, an increase in solute content generally reduces the amount of alpha-phase present. The percent alpha present is also markedly influenced by the thermal and mechanical treatment the delta stabilized alloy has undergone.

The delta stabilized alloys have been considered for certain nuclear applications, such as fuel elements, due to their inherently greater dimensional stability as compared

to alpha-phase materials. The physical and mechanical properties of an alloy as well as its stability depend strongly on the amount of alpha and delta-phase present. It therefore is desirable to have a rapid, economical, non-destructive means of determining the percent alpha in a delta stabilized plutonium alloy.

This paper describes a reliable, nondestructive X-ray method using a diffractometer for quantitatively determining the percent alpha in delta stabilized plutonium alloys.

DEVELOPMENT OF THEORY

The premise of determining the percent alpha in delta stabilized plutonium alloys by X-ray techniques is somewhat fundamental. A sample containing only delta and alpha-phase plutonium would produce a diffraction pattern comprised of all the reflections produced by delta and alpha-phase material separately. In the combined (alpha plus delta) pattern from the two-phase sample, the intensity of the alpha pattern contributing to it, compared to the intensity of the delta pattern contributing to it, depends on the relative proportions of the two-phases present.

The theory behind employing X-ray diffraction in quantitative analysis of conventional materials is established and has been discussed by Klug and Alexander⁽⁴⁾ and Cullity.⁽⁵⁾ The underlying theory is as follows.

Using a single-phase powder specimen of effectively infinite thickness, free from preferred orientation, and having a particle size sufficiently small to enable extinction and micro-absorption effects to be neglected, the diffracted intensity emitted in a diffractometer is given by

$$I = \left(\frac{I_0 e^4}{m^2 c^4} \right) \left(\frac{\lambda^3 A}{32 \pi r} \right) \left(\frac{1}{v^2} \right) \left[|F|^2 p \left(\frac{1 + \cos^2 2\theta}{\sin^2 \theta \cos \theta} \right) \right] \cdot \left(e^{-2M} \right) \left(\frac{1}{2\mu} \right) \quad (1)$$

where e , m , c , λ , F , p , θ and M have their usual meanings.

I = integrated intensity per unit length of diffraction line;

I_0 = intensity of the incident beam,

r = radius of the diffractometer,

A = cross-sectional area of the incident beam,

v = volume of the unit cell,

μ = linear absorption coefficient of the specimen.

(1) Ellinger, F. H., Land, C. C., and Miner, W. N., "Solubility Limits of Aluminum in Delta Plutonium and some Revisions of the Plutonium-Aluminum Phase Diagram," *J. Nuclear Materials*, 5, 165-72 (1962).

(2) Selle, J. E., and Etter, D. E., "Plutonium-Cerium System," *Trans. Am. Inst. Mining Met. Engr.*, 230, 1000-5 (1964).

(3) Ellinger, F. H., Land, C. C., and Struebing, V. O., "Plutonium-Gallium System," *J. Nuclear Materials*, 12, 226-36, (1964).

(4) Klug, H. P., and Alexander, L. E., *X-ray Diffraction Procedure for Polycrystalline and Amorphous Materials*, New York, J. Wiley and Sons, Inc., 1954.

(5) Cullity, B. D., *Elements of X-ray Diffraction*, Reading, Mass., Addison-Wesley Pub. Co., Inc., 1956.

Equation (1) can be simplified to the form,

$$I = kR \frac{1}{2\mu}, \quad (2)$$

where

$$k = \left(\frac{I_0 e^4}{m^2 c^4} \right) \left(\frac{\lambda^3 A}{32\pi r} \right), \quad (3)$$

i.e., k depends on the experimental conditions, and

$$R = \left(\frac{1}{v^2} \right) \left[|F|^2 p \left(\frac{1 + \cos^2 2\theta}{\sin^2 \theta \cos \theta} \right) \right] \left(e^{-2M} \right), \quad (4)$$

i.e., R depends on the unit cell geometry and the set of planes chosen.

For a sample consisting of a uniform mixture of n components, the intensity of the X-rays diffracted by the i^{th} component for a given setting is given by

$$I_i = \frac{kR_i f_i}{2\mu}, \quad (5)$$

where

$$f_i = \frac{x_i / \rho_i}{\sum_1^n (x_i / \rho_i)} = \text{volume fraction}$$

and

$$\mu = \frac{\sum x_i (\mu_i / \rho_i)}{\sum (x_i / \rho_i)} = \frac{\sum x_i (\mu_i^*)}{\sum (x_i / \rho_i)}$$

Re-writing Equation (5) in terms of x_1 , the weight fraction of component 1, and μ_1^* , μ_m^* the respective mass absorption coefficients of component 1 and the remainder of the specimen gives the general relation

$$I_1 = \frac{kR_1 x_1}{\rho_1 [x_1 (\mu_1^* + \mu_m^*) + \mu_m^*]}. \quad (6)$$

This equation is the basic relationship underlying all quantitative diffraction analysis with the X-ray spectrometer. It relates the intensity of a selected diffraction line of the unknown components to its concentration, density, and mass absorption coefficient, and to the mass absorption coefficient of the matrix.

In the present case, we are dealing with a two-phase mixture of alpha + delta. Therefore, Equation (6) reduces to

$$I_\alpha = \frac{kR_\alpha x_\alpha}{\rho_\alpha (x_\alpha \mu_\alpha^* + x_\delta \mu_\delta^*)} \quad (7)$$

A similar expression exists for I_δ .

There are three analytical procedures one can pursue at this point. These are the single line method, the direct comparison method, and the internal standard method. All of these methods have one essential feature in common. The measurement of the concentration of a particular phase depends on the measurement of the ratio of the intensity of a diffraction line from that phase to the intensity of some standard reference line. In the single line method, the reference line is a line from the pure phase. In the direct comparison method, it is a line from another phase in the mixture. In the internal standard method, it is a line from a foreign material mixed with the specimen.

The direct comparison method is used in this investigation since it is the only method that is readily applicable to massive, polycrystalline, metallurgical specimens of varying size. In the present case

$$\mu_\alpha^* \approx \mu_\delta^*,$$

and when the I_α , I_δ ratio is taken, one obtains

$$I = \frac{R_\alpha x_\alpha \rho_\alpha}{R_\delta x_\delta \rho_\delta}.$$

Solving explicitly for w/o α , i.e., 100 x_α , one obtains

$$\text{w/o } \alpha = \frac{100}{1 + \left(\frac{I_\delta}{I_\alpha} \frac{R_\alpha}{R_\delta} \frac{\rho_\alpha}{\rho_\delta} \right)}. \quad (8)$$

This is the basic equation used to determine the w/o alpha present in delta stabilized plutonium alloys.

For ease in handling Equation (8), it is desirable to set

$$\frac{R_\alpha \rho_\alpha}{R_\delta \rho_\delta} = R_t,$$

thus yielding

$$\text{w/o } \alpha = \frac{100}{1 + \frac{I_\delta}{I_\alpha} R_t}. \quad (9)$$

Where

I_δ = Integrated intensity of certain selected delta peaks of the unknown

I_α = Integrated intensity of certain selected alpha peaks of unknown.

DETERMINATION OF CONSTANT

Before Equation (9) can be applied to our problem, it is first necessary to evaluate the constant R_t . This can be

done either experimentally or theoretically. However, due to the uncertainty of dependence of atomic scattering factors on the 2θ angle, size of dispersion correction and the influence of the temperature factor, it is best to use the experimental approach.

Randomly oriented standards of 100 percent alpha and 100 percent delta were prepared. The alpha plutonium was from an as-cast ingot having a total impurity content of approximately 250 ppm. The delta plutonium specimen was from a homogenized Pu-1 w/o Ga alloy having approximately 250 ppm total impurities. The homogenization treatment for the delta material was 72 hours at 475°C under vacuum.

Small particles were obtained from the two materials by filing in an argon atmosphere box. After filing, the particles were sized by sieving. The fraction used was that caught between the 200 and 325 mesh screens. Any iron contamination from the file was removed by passing a magnet through the sized powder. The powders were then placed in individual tantalum crucibles which were in turn placed in glass ampules which were then evacuated to 5×10^{-6} torr and sealed.

The encapsulated alpha powder was stress relieved for 32 hours at 100°C and cooled to room temperature at 4°C/hour. The encapsulated delta powder was stress relieved for 1-1/2 hours at 265°C and cooled at approximately 20°C/hour.

The powders were individually mounted in press-fitting tubular mounts. Mylar film in sheet form (thickness 0.25 mil) was stretched over the smaller tube having dimensions 7/8-in. OD by 25/32-in. ID by 1/2 inch. This was then pressed into the larger tube of dimensions 1-in. OD by 29/32-in. ID by 1/2 inch. The inner side of the sheet had previously been very lightly coated with a hydrocarbon grease. The unit was then placed face down on a clean surface in an open-face hood. The stress relieved powder was then decapsulated and poured into the unit giving a level 1/32-in. layer, 25/32 in. in diameter. A press-fit plug was used to back the powder and maintain the uniform layer thickness. The loose sheet of Mylar was then twisted and taped. The sealed unit was moved to a clean sheet and double bagged in 0.25 mil sheet. The bagged sample was then ready for transfer to a diffractometer.

Diffractometer tracings were obtained for both alpha and delta on the same diffractometer under identical settings. This procedure was duplicated on a separate diffractometer and the results were averaged. The average integrated intensities are listed in Table I. No alpha was detected in the diffractometer patterns of the delta standard.

The strong alpha peak was, as would be expected, the overlapping (020) and (211) reflections. These two

TABLE I

INTERPLANAR SPACINGS, DIFFRACTION ANGLES, AND RELATIVE INTEGRATED INTENSITIES OF ANNEALED ALPHA AND DELTA POWDERS

Alpha Plutonium				Delta Plutonium			
h,k,l	I	$d(\text{\AA})$	θ° ($\text{CuK}\alpha$)	h,k,l	I	$d(\text{\AA})$	θ° ($\text{CuK}\alpha$)
013	0.08	2.873	15.56	111	1.34	2.656	16.86
113	0.36	2.781	16.12	200	0.70	2.302	19.56
201		2.767	16.18	220	0.58	1.630	28.22
004		2.684	16.69	311	0.74	1.390	33.69
104	0.02	2.657	16.87	222	0.20	1.330	35.40
211	0.24	2.601	17.24	400	0.08	1.152	41.96
203		2.586	17.34	331	0.36	1.057	46.78
210		2.564	17.50	420	0.31	1.031	48.36
212	0.02	2.486	18.06	422	0.26	0.941	54.96
020	1.46	2.413	18.63	511,333	0.40	0.8889	60.30
211		2.400	18.74	440	0.08	0.8152	70.88
014		2.345	19.19				
114	0.29	2.329	19.32				
104		2.287	19.69				
213		2.280	19.76				
212	0.04	2.171	20.79				
105		2.166	20.85				
300		2.032	22.30				
302	0.09	2.029	22.33				
312		1.872	24.32				
310		1.866	24.40				
222	0.10	1.855	24.55				
204		1.831	24.90				
106		1.816	25.12				
221	0.12	1.813	25.16				
215		1.808	25.24				
304		1.800	25.36				
313	0.12	1.796	25.42				
024		1.793	25.46				
006		1.791	25.49				
124	0.17	1.789	25.52				
223		1.763	25.93				
222		1.714	26.72				
214	0.16	1.712	26.75				

reflections are used as the alpha reference peak since (1) there are no interfering delta peaks, (2) they are difficult to quantitatively separate, and (3) the additive strength of these two reflections is four times as large as any other alpha peak. This latter effect increases the sensitivity at low alpha contents.

The choice of a reference delta peak is less straightforward. The reason is the degree of preferred orientation possible in worked and/or recrystallized delta-phase alloys. If a single delta peak was to be used, the (311) reflection would be chosen since of the major peaks it

seems to be the least affected by preferred orientation. The first four peaks of delta, i.e., (111), (200), (220), and (311), in diffractometer patterns are generally well defined, readily readable, and of comparable intensities. To minimize the effect of preferred orientation, the reference peaks chosen for delta are the (111), (200), (220), and (311) reflections.

The integrated intensities of the reference alpha and delta peaks in the standard patterns were obtained under identical conditions. This approach allows a calculation of the needed constant for Equation (9).

Since the area of sample bathed in X rays at any given 2θ angle was equal for both of the standards, the area percent alpha was 50 percent. The area percent is assumed equal to the atomic percent since the differing linear absorption coefficients give rise to different depths of penetration in alpha and delta-phase material. Converting this value to w/o alpha using an atomic weight of 236.3 for the Pu-1 w/o Ga alloy yielded w/o alpha = 50.3 percent.

To evaluate the necessary R_f constant for Equation (9), the integrated areas of the reference alpha and delta peaks in the standard patterns were measured. Substituting these values into Equation (9) using the value of 50.3 w/o alpha for the standards, yields $R_f = 0.430$.

DESCRIPTION OF METHOD

SPECIMEN PREPARATION

The percent alpha in delta stabilized alloys is strongly dependent on the thermal and mechanical history of the specimen. This dictates that the sample must be examined in the massive form and that the surface be free from worked induced during sample preparation. The surface should also be relatively free of oxides, hydrides, etc., since these would tend to mask the underlying plutonium reflections.

If the sample is flat and its surface representative of the bulk material or if looking for a surface effect, the sample need only be electropolished in an 8:5:5 solution of orthophosphoric acid, glycerol, and ethyl alcohol at 30 volts for a few seconds to obtain a clean surface. If the sample has undergone any induced work since the desired thermal and/or mechanical treatment (e.g., during sampling), it is necessary to electropolish approximately 4 minutes in the cooled solution to guarantee removal of the worked surface layer. If the sample has undergone a metallographic preparation it is necessary to electropolish for approximately 2 minutes to guarantee removal of the worked surface layer.

The prepared specimen is mounted in a 1-in. OD by 7/8-in. ID by 3/4-in. ring and backed with an Apiezon Q compound

if necessary. The unit is bagged in 0.25-mil sheet of Mylar film, sealed with tape, and transferred to the diffractometer. If the sample is in a metallographic mount, it is only bagged and transferred to the diffractometer.

MAKING THE MEASUREMENT

The experimental procedure for determining the percent alpha is to obtain an X-ray diffractometer pattern of a massive specimen at $1^\circ 2\theta$ per minute using a sample spinner. The sample is scanned from 27° to $70^\circ 2\theta$ using filtered $\text{CuK}\alpha$ radiation and a proportional counter. A pulse height discriminator is used to reduce spurious effects due to fluorescent and white radiation. The integrated intensity above the background level is measured with a planimeter for the (020) and (211) alpha peaks and for the (111), (200), (220), and (311) delta peaks. The delta peaks are corrected for alpha contributions. The w/o alpha is then calculated using Equation (9).

For alpha contents less than 10 w/o, it is necessary to scan the region between 35.5° to $38.5^\circ 2\theta$ at a scale factor magnification of 10, 20, or 30X. For alpha contents less than 4 w/o this region is retraced four times. The measured area under the alpha peak is divided by the increase in scale magnification before use in Equation (9).

It requires about one hour for recording the pattern automatically, measuring the integrated intensities with a planimeter and calculating the results. Greater precision could undoubtedly be obtained, but more time would be used to record the integrated intensities and measure the data. However, the added accuracy is not justified economically, because of the inherent errors of the X-ray methods for determining the percent alpha.

SOURCES OF ERRORS

PROBLEMS OF DIFFRACTION FROM PLUTONIUM

There are three main sources of difficulty encountered in obtaining diffraction patterns from plutonium samples:

High Absorption Coefficient of Plutonium for X Rays -

It is well known that metallurgical specimens strongly absorb X rays and hence the diffracted beam must originate from a thin surface layer. This effect is especially pronounced in plutonium due to its high absorption coefficient for X rays. Roof⁽⁶⁾ has reported that $\mu/\rho = 513 \pm 19 \text{ cm}^2 \text{ g}^{-1}$ (experimental) and $48^{\circ} \text{ cm}^2 \text{ g}^{-1}$ (theoretical) using $\text{CuK}\alpha$ radiation.

(6) Roof, R. B., "X-ray Absorption Coefficients of Thorium, Uranium, and Plutonium - Experimental Determination and Theoretical Interpretation," *Phys. Rev.*, 113, 820-5 (1959).

The results of approximate calculations for the effective depth of penetration for a diffracted beam for plutonium using $\text{CuK}\alpha$ radiation at $45^\circ 2\theta$ and $90^\circ 2\theta$ are shown in Figure 1. At $45^\circ 2\theta$ the calculations yielded values of 1400Å for 50 percent contribution and 6000Å for 95 percent contribution. Thus, in plutonium, the diffracted beam originates from a very thin surface layer. For a discussion of this calculation see Cullity.⁽⁵⁾

The depth of penetration is a function of the wave length of radiation used. For example, the longer wave length chromium radiation would penetrate less than half the distance shown by copper radiation. The shorter wave length molybdenum radiation would penetrate considerably deeper than copper radiation. Hence, to minimize the importance of surface condition of the specimen, one might consider using a shorter wave length such as $\text{MoK}\alpha$. However, this is undesirable for two reasons: (1) the resulting pattern is more compressed making it more difficult to resolve the alpha and delta reflections, and (2) the intensity ratio of characteristic to white radiation is relatively low thus increasing background in the diffractometer pattern. Copper radiation serves as a reasonable compromise since it has: (1) reasonable penetrating power, (2) high intensity output, and (3) easy availability.

(5) Cullity, B. D., *loc. cit.*, p 1 (p 269).

Figure 1. THE FRACTION G_x OF THE TOTAL DIFFRACTED INTENSITY CONTRIBUTED BY A SURFACE LAYER OF DEPTH x , FOR PLUTONIUM USING $\text{CuK}\alpha$ RADIATION

The solution to the problem of low penetration is to make the surface region representative of the bulk sample. This is accomplished by imparting a work free metallographic finish to the specimen. The method used to attain a work free surface is described in the procedure section.

The importance of achieving a work free surface cannot be over emphasized.

High Reactivity of Plutonium with the Atmosphere -

This results in a surface layer (primarily and ordinarily PuO_2) having a high absorption coefficient for X rays. A complicating factor would be preferential oxidation of the two phases. This could result in a preferential masking of one of the constituents and erroneous results.

The obvious solution to this problem is to restrict the thickness of the oxide layer. This is readily accomplished by electropolishing the specimen in a cooled solution followed by examination within a few hours after preparation. The absence of oxide reflections in the diffractometer patterns of metallographically prepared alpha and delta specimens is verification that this effect can be made insignificant.

Self Radiation of Plutonium Specimens -

The self radiation of plutonium can produce spurious effects in counter measurements. The effect leads to an increase in background in the resulting diffractometer patterns. The solution to this problem is to use a pulse-height discriminator. The single-channel, pulse-height analyzer is set to pass only pulses due to $\text{CuK}\alpha$ radiation and to reject those due to other wave lengths. The pulse-height discriminator also reduces the spurious effects due to fluorescent and white radiation.

ERRORS PECULIAR TO THE METHOD

There are certain effects which can cause great difficulty in quantitative analysis because they cause observed intensities to depart widely from the theoretical. The most important of these complicating factors are:

Microabsorption -

For a discussion of the microabsorption effect see Cullity.⁽⁵⁾ For the case in question, the microabsorption effect is negligible since $\mu_a^* \approx \mu_\delta^*$. In addition, the low penetration of the X-ray beam minimizes any affect due to difference in particle size of the alpha and delta phases.

Extinction -

All real crystals are imperfect, in the sense that they have a mosaic structure, and the degree of imperfection can vary greatly from one crystal to another. Equation (1) is derived on the basis of the so-called "ideally imperfect" crystal, one in which the mosaic blocks are quite small (of the order of 10^{-4} to 10^{-5} cm in thickness) and disoriented so that they are all essentially nonparallel. Such a crystal has maximum reflecting power. A crystal made up of large mosaic blocks, some or all of which are accurately parallel to one another, is more nearly perfect and has a lower reflection power. This decrease in the intensity of the diffracted beam as the crystal becomes more nearly perfect is called extinction. Extinction is absent for the ideally imperfect crystal, and the presence of extinction invalidates Equation (1).

For the case in question, extinction is negligible because of the very nature of the formation of alpha in delta. The change in specific volume accompanying the transformation of delta to alpha sets up nonuniform strains in both phases so severe that both kinds of crystals can be considered highly imperfect.

Preferred Orientation -

The basic intensity equation, Equation (1), is derived on the premise of random orientation of the constituent crystals in the sample and is not valid if any preferred orientation exists. For a solid polycrystalline aggregate, the analyst has no control over the distribution of orientation in it, and should be aware of the possibility of error due to preferred orientation. When the $I_\delta(111)/I_\delta(200)$ deviates from 1.88, preferred orientation exists in the delta matrix. The use of four delta peaks (111, 200, 220, 311) and two alpha peaks (020, 211) minimizes the effect of preferred orientation. Thus, the method is somewhat applicable to specimens exhibiting preferred orientation.

VERIFICATION OF METHOD

To verify the method, samples of known and unknown percent-alpha compositions were examined.

SAMPLES OF KNOWN COMPOSITION

Two massive samples of known percent-alpha composition were prepared and examined, these were: (1) a 0.8 w/o alpha specimen, and (2) a 1.6 w/o alpha specimen.

The 0.8 w/o Alpha Plutonium Specimen -

This test specimen was prepared by inserting a 30-mil alpha wire into a hole drilled in an alpha-free, delta-stabilized, Pu-1 w/o Ga alloy. The delta plutonium was high purity (300 ppm total impurities) and had previously been heat treated 72 hours at 500°C followed by cooling at 20°C/hr. to insure absence of alpha plutonium. The sample was given a one-micron metallographic finish, then electropolished. An X-ray examination showed no evidence of alpha or of preferred orientation. The alpha material was a drawn wire having a diameter of 30 mils. The alpha wire was pressed into an off-center hole drilled in the delta material. The composite surface was then given a short electropolish.

The composite sample was completely bathed in X rays and examined for alpha content using the prescribed procedure. The X-ray method yielded 0.8 w/o alpha, which compares very favorably with the known alpha content of 0.8 ± 0.1 w/o alpha.

The 1.6 w/o Alpha Plutonium Specimen -

This sample was prepared in a similar manner to that described above for the 0.8 w/o alpha specimen. The X-ray method yielded 1.5 w/o alpha, which compares favorably with the known alpha content of 1.6 ± 0.2 w/o alpha.

The good agreement between the known and experimentally determined values verifies the herein prescribed X-ray method for determining low percentages of alpha in delta stabilized plutonium alloys.

SAMPLES OF UNKNOWN COMPOSITION

Specimens of delta stabilized Pu-1 w/o Ga were given various treatments to vary the alpha content. The percent-alpha by X-ray determination and the gravimetric densities of these specimens are shown in Figure 2. As would be

(5) Cullity, B. D., *loc. cit.*, p 1 (p 399).

Figure 2. GRAVIMETRICALLY DETERMINED DENSITIES OF UNKNOWN VS. w/o ALPHA CONTENT AS DETERMINED BY X-RAY MEANS. The solid line is a plot of Equation (10)

expected, there is a direct correlation between alpha content and density.

By assuming a linear density-percent alpha relationship, one can readily derive the expression

$$\rho = 0.0398 \text{ w/o } \alpha + 15.72 \quad (10)$$

The expression being based on experimentally determined densities of 19.70 g/cm³ for microcrack-free alpha and 15.72 g/cm³ for the alpha-free, delta-stabilized Pu-1 w/o Ga alloy. The total impurity content of both materials was approximately 1250 ppm including 300 ppm iron and 60 ppm nickel.

The solid line in Figure 2 is a plot of Equation (10). The agreement between percent-alpha determined by X-ray and by density measurements as shown in Figure 2 is satisfactory. This agreement supports the validity of this prescribed X-ray method for determining percent-alpha in delta stabilized alloys.

DISCUSSION

The method as described is explicitly applicable only to the Pu-1 w/o Ga alloy and has only been verified for low

percentages of alpha. However, the method is readily extended to other compositions and alloy systems without additional experimental work. In fact, the method may be used as is for other compositions and systems without introducing considerable error.

For the method to explicitly apply to other compositions and systems, it is necessary to correct the delta density used in the R_t constant evaluation. Also, it would be necessary to correct for the change in absorption coefficient as a function of solute content in delta.

Before the method could be used with any certainty at high percentages of alpha content, it would first be necessary to experimentally verify the method. An error in the R_t constant would be less noticeable at low percents of alpha than it would at the 50-50 compositions. This dictates that the R_t constant be known accurately for the 50-50 compositional range.

Another problem at higher alpha contents is the subtraction of the alpha contribution to the reference delta peaks. This correction, though time consuming, can be readily made. If random orientation exists, it is possible to use the (220) as the reference delta peak, thus eliminating the alpha contribution correction. This is possible since the alpha peaks that are superimposed on the (220) peak are of weak intensity.

One way to (1) verify the existing R_t constant for all compositions, (2) re-evaluate the R_t constant as a function of percent alpha, and (3) eliminate the need for subtracting out interfering lines would be as follows: Prepare 10 delta and 10 alpha slabs having random orientations and dimensions of 0.020 by 0.20 by 0.20 in. Using 10 slabs at a time it would be possible to canvas the whole range of compositions. A plot of measured I_α/I_δ of certain selected peaks versus known w/o alpha content would yield a relationship from whence the alpha content could be determined from unknowns.

As discussed earlier, the prescribed method is somewhat applicable to materials exhibiting preferred orientation. It would be possible to eliminate the problem of preferred orientation by following a procedure outlined by Lopata and Kula.⁽⁷⁾ This procedure was not used in this investigation due to the amount of work required to make the correction.

(7) Lopata, S. L., and Kula, E. B., "Quantitative Phase Analysis in Textured Materials," *Trans. Am. Inst. Mining Met. Engr.*, 233, 288-93 (1965).

RFP-708