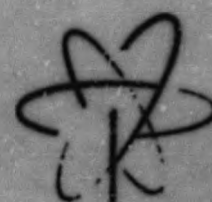


GEAP-4303  
JULY 15, 1963

EURAEC  
JOINT U.S.-EURATOM RESEARCH  
AND DEVELOPMENT PROGRAM

755 ✓



MASTER

# PROGRAM FOR THE DEVELOPMENT OF PLUTONIUM RECYCLE FOR USE IN LIGHT WATER MODERATED REACTORS

## NINTH QUARTERLY REPORT APRIL 1 - JUNE 30, 1963

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U.S. ATOMIC ENERGY COMMISSION  
CONTRACT AT(04-3)-189  
PROJECT AGREEMENT 21

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GENERAL  ELECTRIC

ATOMIC POWER EQUIPMENT DEPARTMENT  
SAN JOSE, CALIFORNIA

EURAEC  
GEAP-4303  
Joint U.S.-EURATOM Research  
and Development Program

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AEC Contract AT(04-3)-189, P. A. No. 21  
for the  
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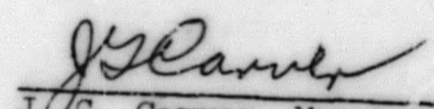
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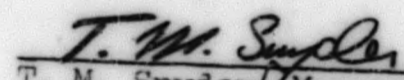
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### INTRODUCTION

The United States and the European Atomic Energy Community (EURATOM) on May 29, and June 18, 1958, signed an agreement which provides a basis for cooperation in programs for the advancement of the peaceful applications of atomic energy. This agreement, in part, provides for the establishment of a Joint U.S.-EURATOM research and development program which is aimed at reactors to be constructed in Europe under the Joint Program.

The work described in this report represents the Joint U.S.-EURATOM effort which is in keeping with the spirit of cooperation in contributing to the common good by the sharing of scientific and technical information and minimizing the duplication of effort by the limited pool of technical talent available in Western Europe and the United States.

## I. SUMMARY

### 1.1 Statement of Problem

The objective of this program is the experimental determination of basic physical data and employment of these data to establish and confirm a theoretical model for predicting the long-term changes in isotopic composition and reactivity in plutonium-enriched, light-water-moderated reactors. Such a model is required for the sound economic evaluation of any proposed design for a light-water reactor using plutonium enrichment, particularly any reactor using plutonium recycle.

The required data are being obtained from a burnup experiment in which a portion of a uranium-plutonium fuel lattice, typical of light-water-moderated reactors, is being irradiated to high burnup in the Vallecitos Boiling Water Reactor. The technique of resonance activation is being used to provide, at several locations within the fuel element, experimental verification of the calculated slow neutron spectrum. Changes in isotopic composition as a function of fuel burnup are determined by mass spectroscopic assay of fuel samples extracted at appropriate irradiation times.

The need for such burnup and activation data is discussed at length in a previous Quarterly, GEAP-4081, as is the basic approach employed in the Program. The model which is sought may be considered as a set of procedures, grounded in all of the known relevant physical data and theory, which is capable of accurate prediction both of the slow neutron spectra within the Program fuel element and of the observed changes in isotopic composition with burnup.

### 1.2 Quarterly Progress

The project fuel element received an estimated burnup of 831 MWD/T during the quarter, which brings the total to 3165 MWD/T. This exposure is as logged by VBWR operating personnel. Applying the same scale factor between logged exposure and Ce-Cs analysis of the first fuel sample gives a corrected exposure of 3774 MWD/T.

The differential solution experiment indicated that less than 1% of the contained plutonium occurred as segregated pure  $\text{PuO}_2$ . The "hot spots" observed via autoradiography of polished samples of fuel indicated that a substantial fraction of the  $\text{PuO}_2$  is contained in the hot spots.

Super-Ortho-Press plates were used to make enlargements of autoradiographs of polished unirradiated fuel pellets. Densitometer measurements were made on the enlargements. It was observed that the contrast range on the autoradiographs were too wide to allow the SOP plates to be exposed in the linear region of the D-log E curve for both the hot spots and the solid solution simultaneously. However, a five-minute  $\alpha$ -exposure of a large hot spot gave the same darkening as a 480-minute  $\alpha$ -exposure of the background, from which it is possible to infer a corrected value of 82 times for the ratio of the Pu density in the hot spot to that in the background.

Further debugging of EPITHERMØS, the epithermal extension of the BNL THERMØS code, has been made and a test problem has been run.

### 1.3 Administrative

All program efforts were temporarily suspended during the first half of the quarter except for continued irradiation in the VBWR. The suspension was in compliance with instructions received from the San Francisco Operations Office.

### 1.4 Principal Investigators

M. A. Robkin of General Electric Atomic Power Equipment Department and  
M. P. Lagache of EURATOM.

## II. DESCRIPTION OF WORK - EXPERIMENTAL

### 2.1 Fuel Irradiation

The project fuel element received an estimated burnup of 831 MWD/T during the quarter, which brings the total to 3165 MWD/T. This exposure is as logged by VBWR operating personnel. Applying the same scale factor between logged exposure and Ce-Cs analysis of the first fuel sample gives a corrected exposure of 3774 MWD/T.

### 2.2 Differential Solution

It has been observed by various investigators (see, for example, J. D. L. Harrison in The Fourth Plansee Seminar on "Powder Metallurgy in the Nuclear Age") that  $\text{UO}_2$  enriched to over 60% in  $\text{PuO}_2$  will not dissolve in dilute nitric acid whereas enrichments below this value dissolve readily. Advantage was taken of this difference of solubility to determine the amount of  $\text{PuO}_2$  in the project fuel which occurs in concentrations above 60%.

Starting with an initial dissolving medium of 100 ml of 8M.  $\text{HNO}_3$ , the time of solution and the temperature of the solvent were increased stepwise. Samples were obtained at each step by withdrawing 25 microliters of solution and diluting to 10 ml from which 25 microliter mounts were prepared in triplicate for Pu alpha counting. Following dissolution at 8M., the contents of the flask were filtered through a glass filter. The filter and undissolved material were returned to the flask and dissolution continued in 50 ml of 16M.  $\text{HNO}_3$ , samples again being obtained as before. Samples 11 and 12 were .005M. and .010M. in HF, respectively. The calculated percent of Pu dissolved is based on the total alpha count encountered in sample 6 plus sample 12. The results are displayed in Table 2.2.1.

TABLE 2.2.1 - RELATIVE DISSOLUTION OF CONTAINED PLUTONIUM

| Sample No. | Conditions                        | Approx. % Pu<br>Dissolved |
|------------|-----------------------------------|---------------------------|
| 1          | 8M., 25°, 1/2 hr.                 | 34.6                      |
| 2          | 8M., 25°, 2 hr.                   | 71.6                      |
| 3          | 8M., 60°, 1/2 hr.                 | 97.1                      |
| 4          | 8M., 60°, 2 hr.                   | 97.6                      |
| 5          | 8M., 100°, 1/2 hr.                | 99.4                      |
| 6          | 8M., 100°, 2 hr.                  | 99.2                      |
| 7          | 16M., 25°, 1/2 hr.                | .51                       |
| 8          | 16M., 25°, 2 hr.                  | .56                       |
| 9          | 16M., 100°, 1/2 hr.               | .51                       |
| 10         | 16M., 100°, 2 hr.                 | .55                       |
| 11         | 16M., .005 M.HF,<br>120°, 1/2 hr. | .84                       |
| 12         | 16M., .010 M.HF,<br>120°, 4 hr.   | .76                       |

As can be seen from the experimental results, there did not seem to be a significant increase in plutonium alpha activity when the acid concentration was increased to 16 Molar. The implication is that there is less than 1% of the contained plutonium occurring as segregated pure  $\text{PuO}_2$ .

In order to reconcile these results with the autoradiograph results described below, it is necessary to postulate that sufficient diffusion occurred during the sintering process to reduce the  $\text{PuO}_2$  concentration in the segregated lumps to a value below 60%. In this case, the resulting lumps would show a solubility behavior similar to that of  $\text{UO}_2$  with low enrichment of  $\text{PuO}_2$ .

### 2.3 Autoradiography

Determination of plutonium concentrations by means of photography is based on the characteristic curve for sensitive emulsions. This curve has the appearance of Figure 2.3.1.

In the Figure,  $I_0$  is the transmission intensity of a light beam through unblackened film, and  $I$  is the transmission through film blackened due to an exposure  $E$  (meter-candle-sec).

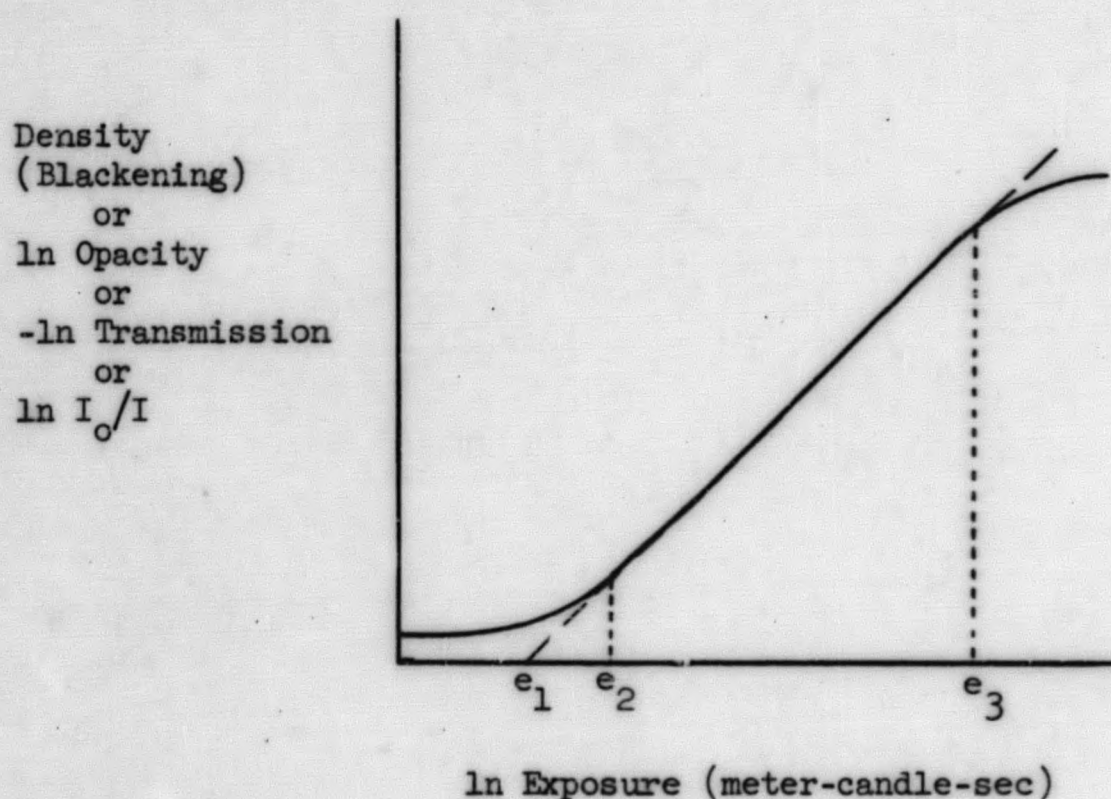


Figure 2.3.1 - Typical Exposure Curve  
for Photographic Emulsions

In order to interpret optical enlargements of autoradiographs, it is necessary that the exposures be made in the straight line part of the curve. To determine the slope of the autoradiograph emulsion and of the enlargement emulsion, we proceed as follows:

Consider a point on the fuel surface of intensity  $A$  (arbitrary units of energy/time). Expose the first emulsion (autoradiograph) for the times  $t_1$ ,  $t_2$ . The darkening in the straight line part of the curve may be taken as

$$\begin{aligned} D_1 &= \gamma \ln t_1 A \\ D_2 &= \gamma \ln t_2 A \end{aligned} \quad (2.3.1 \text{ a,b})$$

where we neglect the pre-sensitizing exposure,  $e_1$ .

The light transmitted through this emulsion during the enlargement exposure is given by

$$T = I_o e^{-D} \quad (2.3.2)$$

where  $I_0$  is the intensity of light transmitted through the undarkened emulsion.

Thus,

$$T_1 = \frac{I_0}{(t_1 A)^\gamma}$$

$$T_2 = \frac{I_0}{(t_2 A)^\gamma}$$

(2.3.3 a,b)

A second emulsion is exposed (enlargement) for two times,  $\Theta_1$ , with each of the previous two images. The densities resulting are given by

$$d_{11} = \beta \ln T_1 \Theta_1 = \beta \ln \frac{I_0 \Theta_1}{(t_1 A)^\gamma}$$

$$d_{12} = \beta \ln T_1 \Theta_2 = \beta \ln \frac{I_0 \Theta_2}{(t_1 A)^\gamma}$$

(2.3.4 a,b,c,d)

$$d_{23} = \beta \ln T_2 \Theta_3 = \beta \ln \frac{I_0 \Theta_3}{(t_2 A)^\gamma}$$

$$d_{24} = \beta \ln T_2 \Theta_4 = \beta \ln \frac{I_0 \Theta_4}{(t_2 A)^\gamma}$$

Reading these images with a densitometer we obtain transmissions defined as

$$R = J_0 e^{-d}$$

where  $J_0$  is again the intensity reading in the case of no darkening.

We have

$$R_{11} = J_0 \frac{(t_1 A)^{\beta\gamma}}{(\Theta_1 I_0)^\beta}$$

$$R_{12} = J_0 \frac{(t_1 A)^{\beta\gamma}}{(\Theta_2 I_0)^\beta}$$

(2.3.5 a,b,c,d)

$$R_{23} = J_0 \frac{(t_2 A)^{\beta\gamma}}{(\Theta_3 I_0)^\beta}$$

$$R_{24} = J_0 \frac{(t_2 A)^{\beta\gamma}}{(\Theta_4 I_0)^\beta}$$

Thus

$$\frac{R_{11}}{R_{12}} = \left( \frac{\Theta_2}{\Theta_1} \right)^\beta$$

$$\frac{R_{23}}{R_{24}} = \left( \frac{\Theta_4}{\Theta_3} \right)^\beta$$

$$\frac{R_{11}}{R_{23}} = \left( \frac{t_1}{t_2} \right)^{\beta\gamma} \left( \frac{\Theta_3}{\Theta_1} \right)^\beta$$

$$\frac{R_{12}}{R_{24}} = \left( \frac{t_1}{t_2} \right)^{\beta\gamma} \left( \frac{\Theta_4}{\Theta_2} \right)^\beta$$

or

$$\beta = \frac{\ln \frac{R_{11}}{R_{12}}}{\ln \frac{\Theta_2}{\Theta_1}} = \frac{\ln \frac{R_{23}}{R_{24}}}{\ln \frac{\Theta_4}{\Theta_3}} \quad (2.3.6 \text{ a,b})$$

and

$$\gamma = \frac{\frac{1}{\beta} \ln \frac{R_{11}}{R_{23}} - \ln \left( \frac{\Theta_3}{\Theta_1} \right)}{\ln \left( \frac{t_1}{t_2} \right)} = \frac{\frac{1}{\beta} \ln \frac{R_{12}}{R_{24}} - \ln \left( \frac{\Theta_4}{\Theta_2} \right)}{\ln \left( \frac{t_1}{t_2} \right)} \quad (2.3.7 \text{ a,b})$$

The result obtained from eqn. (2.3.6 a) must agree with that from eqn. (2.3.6 b) and, similarly the result from eqn. (2.3.7 a) must agree with that from (2.3.7 b) or else the exposures were not in the linear range. This must be true for all points on the emulsions used in the analysis.

We select another point on the emulsion corresponding to another point on the fuel surface of intensity B (arbitrary units of energy/time).

We find

$$S_{11} = J_0 \frac{(t_1 B)^{\beta\gamma}}{(I_0 e_1)^\beta}$$

$$S_{12} = J_0 \frac{(t_1 B)^{\beta\gamma}}{(I_0 e_2)^\beta}$$

$$S_{23} = J_0 \frac{(t_2 B)^{\beta\gamma}}{(I_0 e_3)^\beta}$$

$$S_{24} = J_0 \frac{(t_2 B)^{\beta\gamma}}{(I_0 e_4)^\beta}$$

(Where the  $S_{ij}$  are constructed in the same manner as the  $R_{ij}$ , but with source strength B instead of A.)

Having already determined  $\beta$  and  $\gamma$ , we find

$$B/A = \left( \frac{S_{11}}{R_{11}} \right)^{1/\beta\gamma}$$

$$= \left( \frac{S_{12}}{R_{12}} \right)^{1/\beta\gamma}$$

$$= \left( \frac{S_{23}}{R_{23}} \right)^{1/\beta\gamma}$$

$$= \left( \frac{S_{24}}{R_{24}} \right)^{1/\beta\gamma}$$

(2.3.8 a,b,c,d)

A check on the consistency of the experiment may be obtained by comparing the ratio B/A obtained from eqns. (2.3.8 a,b,c, and d).

### 2.3.1 Correction for Toe of Exposure Curve

If the density measurements are limited to values in the linear part of the curve with the exposure E in the range  $e_2 \leq E \leq e_3$ , we may extrapolate

the density curve to zero at exposure  $e_1$  and write for the autoradiograph

$$\begin{aligned} D &= \gamma (\ln E - \ln e_1) \\ &= \gamma \ln E' \end{aligned}$$

where

$$E' = tA/e_1 \equiv t'A$$

Similarly for the enlargement we write

$$\begin{aligned} d &= \beta \left[ \ln \frac{I_o^\Theta}{(t'A)^\gamma} - \ln f_1 \right] \\ &= \beta \ln \frac{I_o^{\Theta'}}{(t'A)^\gamma} \end{aligned}$$

where  $f_1$  is a similar extrapolated intercept for the second emulsion.

Since the  $t$ 's and  $\Theta$ 's always appear as ratios in the equations, the results are not affected by the constant exposure corrections. This is true so long as the exposures are restricted to the linear range.

### 2.3.2 Some Experimental Results

Super-Ortho-Press plates were exposed in two series. The five-minute  $\alpha$ - $\beta$  autoradiograph of a fuel pellet irradiated to 1860 MWD/T was enlarged to 100 diameters and a region around a hot spot was photographed for 1/5 sec. and 1/2 sec. on SOP plates. The same position was selected on the 10-minute  $\alpha$ - $\beta$  autoradiograph and another 100 X enlargement was made with 4-sec. and 10-sec. exposures. It was expected that the different exposure times would compensate for the large differences in blackening between the 5-min. and the 10-min. autoradiograph.

The SOP plates were scanned with a Jarrell-Ash Microphotometer. Several locations were selected on one of the plates and the other plates were marked so that a reading could be taken at the same points on the other three plates. The microphotometer was adjusted so that the galvanometer read approximately full scale when the least dense point of the least exposed SOP plate was over the aperture of the densitometer. We denote

readings of the 5-min./ 1/5 sec. plate by  $R_{11}^i$ , the 5-min./1/2 sec. plate by  $R_{12}^i$ , the 10-min./4 sec. plate by  $R_{23}^i$  and the 10-min./10 sec. plate by  $R_{24}^i$  where  $i$  denotes the point on each plate being scanned. The readings obtained are displayed in Table 2.3.2.1.

TABLE 2.3.2.1  
DENSITOMETER READINGS OF AUTORADIOGRAPH ENLARGEMENTS

| Point | $R_{11}^i$ | $R_{12}^i$ | $R_{23}^i$ | $R_{24}^i$ | $\beta$<br>(2.3.6.a) | $\beta$<br>(2.3.6.b) |
|-------|------------|------------|------------|------------|----------------------|----------------------|
| 1     | 97.65      | 93.28      | 67.23      | 32.93      | .05                  | .24                  |
| 2     | 91.7       | 65.6       | 53.1       | 25.0       | .37                  | .25                  |
| 3     | 21.45      | 6.63       | 3.23       | .600       | 1.3                  | .56                  |
| 4     | 24.43      | 7.5        | 3.65       | .925       | 1.3                  | .46                  |
| 5     | 10.85      | 2.48       | -          | -          | 1.6                  | -                    |

As can be seen from the values obtained for  $\beta$ , the SOP emulsion exposure was not confined to the linear range of the characteristic curve. The contrast range of the autoradiograph is extreme; and several different combinations of exposure times,  $\Theta$ , were tried unsuccessfully in an effort to produce a linear response.

Several samples of unirradiated fuel pellets were polished and  $\alpha$  autoradiographs were taken for various times. By reading the autoradiograph directly with the microphotometer with a reduced aperture, one emulsion is eliminated. It is hoped that the  $\alpha$  exposure will result in an exposure in the linear portion of the characteristic curve. Work on these autoradiographs is still in progress.

#### 2.4 Metallography

In order to determine the size and number of the hot spots, a sequence of autoradiographs was taken. One pellet from the fuel pin irradiated to 1860 MWD/T, and three unirradiated fuel pellets were polished. Kodak high resolution photographic slides were exposed. The  $\alpha$ - $\beta$  exposure of the irradiated pellet was made for 5 min. and 10 min., and the  $\alpha$  exposure of the three unirradiated pellets was made for 16 hours with a 1/4-mil mylar shield between the pellet and the emulsion to prevent plutonium contamination of the autoradiograph.

Figures 2.4.1 - 2.4.4 show enlarged views of the autoradiographs. Figure 2.4.1 is an enlargement of the  $\alpha$ - $\beta$  autoradiograph and Figures 2.4.2 - 2.4.4 are the enlargements of the  $\alpha$  autoradiographs.

Since the  $\beta$  range is much larger than the  $\alpha$  range, about 230 microns in  $\text{UO}_2$  and about  $1.7 \times 10^3$  microns in the emulsion (based on  $\text{H}_2\text{O}$ ), it is possible for the  $\beta$ 's to traverse the emulsion at large angles. As a result, the hot spots appear to be somewhat larger than their actual size ( $\beta$  haloing). For this reason, only the  $\alpha$  autoradiographs are considered for the estimation of spot sizes, while the  $\beta$  exposure gives a check on the estimate of the spot density (number of hot spots per unit volume).

It must be pointed out that with only four exposures, it is not possible to obtain a sample of spots which is statistically completely meaningful. The total volume scanned in each pellet by means of the  $\alpha$  exposure is only  $10^{-3} \text{ cm}^3$ , and only  $2.3 \times 10^{-2} \text{ cm}^3$  by means of the  $\beta$  exposure. Since the pellets are about  $1 \text{ cm}^3$  each, we observe a combined total of only  $2.6 \times 10^{-2} \text{ cm}^3$  in the four autoradiographs, or 2.3% of a single pellet volume in the  $\beta$  exposures, and only 0.1% in the  $\alpha$  exposures.

Nevertheless, the protrusion of hot spots is indicative of what may be expected throughout the pellets. A survey of the  $\alpha$  plates gives a spot size distribution as follows.

TABLE 2.4.1 - NUMBER OF HOT SPOTS OBSERVED  
ON THREE POLISHED FUEL PELLET SURFACES

| <u>Diameter (Mils)</u> | <u>Number</u> |
|------------------------|---------------|
| 2                      | 67            |
| 3                      | 31            |
| 4                      | 22            |
| 5                      | 7             |
| 7                      | 8             |
| 10                     | 2             |
| 12                     | 1             |
| 15                     | 2             |
| 17                     | 1             |
| 20                     | 1             |
| 30                     | 1             |

We assume that a section taken through the pellets at random will show, on the average, an identical spot distribution. With this assumption, the distribution displayed in Table 2.4.1 gives a total spot volume of  $.00903 \text{ cm}^3/\text{pellet}$ . The pellet has a volume of  $.924 \text{ cm}^3$ , so the hot spots occupy about 1% of the pellet. We will consider the effect of the hot spots on the irradiation experiment in Sections 3.1 and 3.2.

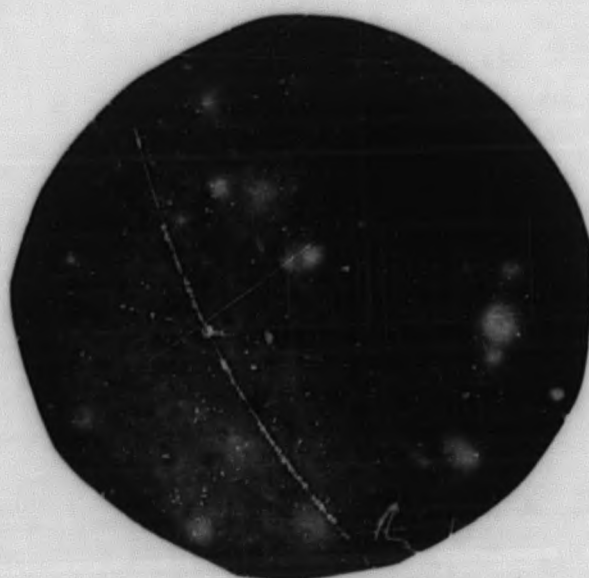


Figure 2.4.1 Autoradiograph of Polished  
Irradiated Fuel Pellet - 6X

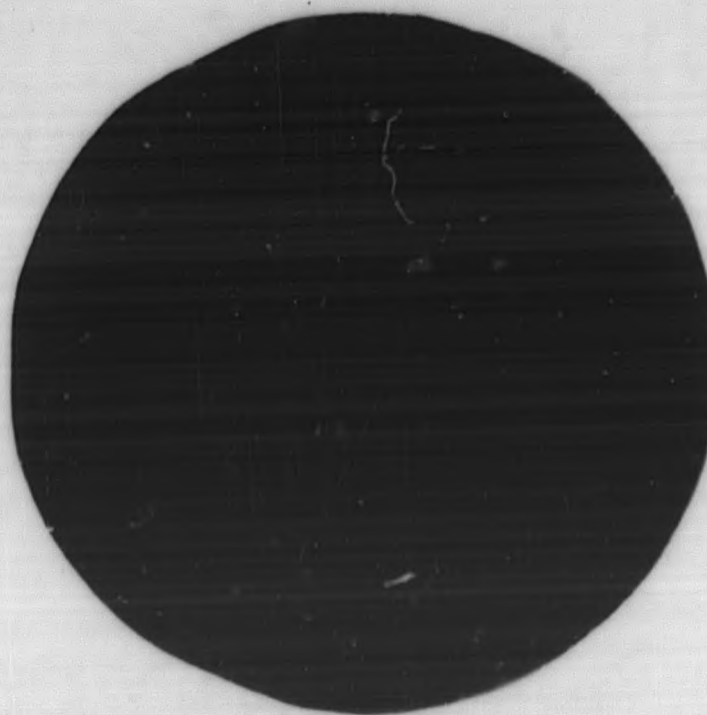


Figure 2.4.2 Autoradiograph of Polished  
Unirradiated Fuel Pellet - 8X

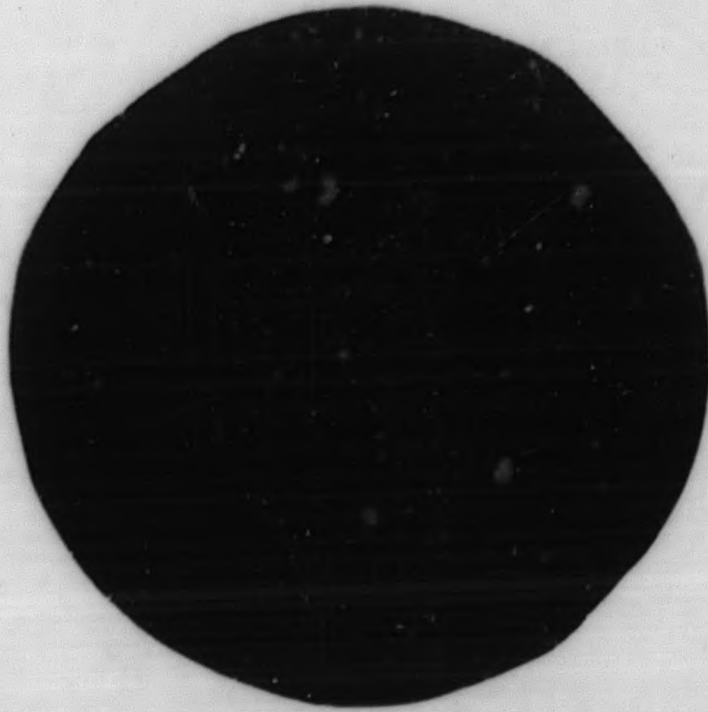


Figure 2.4.3 Autoradiograph of Polished  
Unirradiated Fuel Pellet - 8X

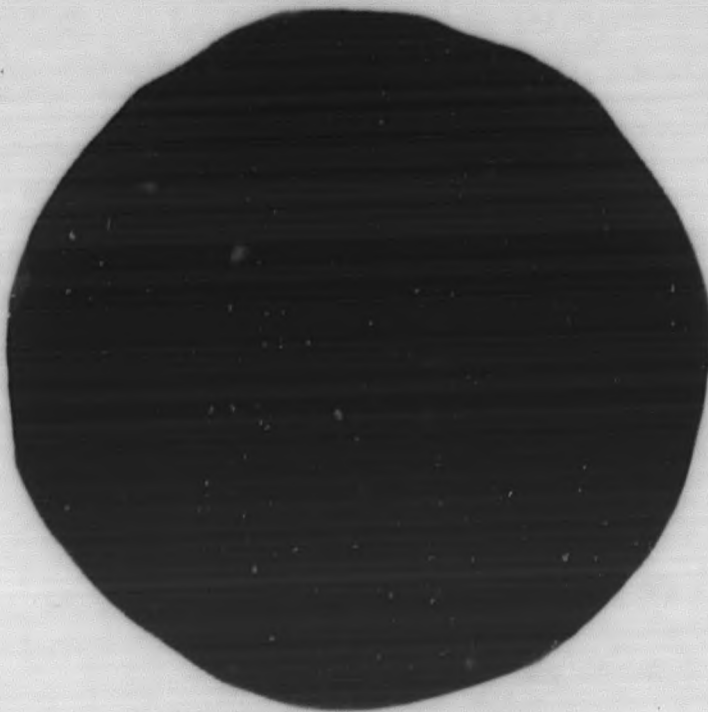


Figure 2.4.4 Autoradiograph of Polished  
Unirradiated Fuel Pellet - 8X

### III. DESCRIPTION OF WORK - THEORETICAL

#### 3.1 Effect of Microsegregation on Isotopic Analysis

The most serious effect of the microsegregation of plutonium is the introduction of an error (due to neglect of self-shielding) into the effective reaction cross sections computed on the basis of a homogeneous solid solution. Since the hot spots seem to occur randomly both as to location and size, it is impossible to obtain a correction which will allow them to be taken into account exactly.

We may illustrate the effect of an error in the reaction cross section by considering the effect on the Pu-240 concentration at any point in the fuel due to an error in the Pu-239 and Pu-240 reaction cross sections.

Assuming that the product of the microscopic cross section times the flux does not change in time, the change in the Pu-239 and the Pu-240 concentrations as a function of time is given by

$$\Delta N^{49} = \left( N_o^{49} - \frac{\sigma_a^{28}}{\sigma_a^{49}} N_o^{28} \right) \left[ 1 - \exp(-\sigma_a^{49} \Theta) \right] \quad (3.1.1)$$

$$\Delta N^{40} = \left( N_o^{40} - \gamma K_1 \right) \left[ 1 - \exp(-\sigma_a^{40} \Theta) \right] - \gamma K_2 \left[ \exp(-\sigma_a^{40} \Theta) - \exp(-\sigma_a^{49} \Theta) \right] \quad (3.1.2)$$

where  $\gamma = \sigma_c^{49} / \sigma_a^{49}$ ,  $\Theta = \text{fluxtime} = \phi t$

$$K_1 = \frac{\sigma_a^{28} N_o^{28}}{\sigma_a^{40}}$$

$$K_2 = \frac{\sigma_a^{49} N_o^{49} - \sigma_a^{28} N_o^{28}}{\sigma_a^{49} - \sigma_a^{40}}$$

The error made in evaluating eqns. (3.1.1) and (3.1.2) due to an error in the cross sections  $\sigma_a^{49}$ ,  $\sigma_a^{40}$  is given by

$$d\Delta N^{49} = \left[ \frac{\sigma_a^{28} N_o^{28}}{\sigma_a^{49}} [1 - \exp(-\sigma_a^{49} \Theta)] + \Theta (\sigma_a^{49} N_o^{49} - \sigma_a^{28} N_o^{28}) \exp(-\sigma_a^{49} \Theta) \right] \frac{d\sigma_a^{49}}{\sigma_a^{49}} \quad (3.1.3)$$

$$\begin{aligned} d\Delta N^{40} = & \left[ (N_o^{40} - \gamma K_1) \sigma_a^{40} \Theta \exp(-\sigma_a^{40} \Theta) + \gamma K_1 [1 - \exp(-\sigma_a^{40} \Theta)] + \gamma K_2 \sigma_a^{40} \Theta \exp(-\sigma_a^{40} \Theta) \right. \\ & \left. - \frac{\gamma K_2 \sigma_a^{40}}{\sigma_a^{49} - \sigma_a^{40}} [\exp(-\sigma_a^{40} \Theta) - \exp(-\sigma_a^{49} \Theta)] \right] \frac{d\sigma_a^{40}}{\sigma_a^{40}} \\ & - \gamma \left[ K_2 \sigma_a^{49} \Theta \exp(-\sigma_a^{49} \Theta) + \frac{\sigma_a^{49} (N_o^{49} - K_2)}{\sigma_a^{49} - \sigma_a^{40}} [\exp(-\sigma_a^{40} \Theta) - \exp(-\sigma_a^{49} \Theta)] \right] \frac{d\sigma_a^{49}}{\sigma_a^{49}} \end{aligned} \quad (3.1.4)$$

The irradiation experiment is designed to reach an exposure of  $10^4$  MWD/Tonne which is equal to about  $3 \times 10^{20}$  fissions/cm<sup>3</sup> on the basis of  $3.1 \times 10^{10}$  fissions/watt-sec for uranium fission and assuming that the energy released in a plutonium fission is about the same as that from a uranium fission. We may approximate the flux time by considering that the initial concentrations are:

$$\begin{aligned} N_o^{25} &= 1.686 \times 10^{-4} \text{ (b-cm)}^{-1} & N_o^{49} &= 2.75 \times 10^{-4} \text{ (b-cm)}^{-1} \\ N_o^{28} &= 2.29 \times 10^{-2} \text{ (b-cm)}^{-1} & N_o^{40} &= 5.79 \times 10^{-5} \text{ (b-cm)}^{-1} \\ & & N_o^{41} &= 9.68 \times 10^{-6} \text{ (b-cm)}^{-1} \end{aligned}$$

The burnup amounts to about 3/5 of the fissionable atoms initially present. Taking the average concentration to be 70% of the initial value, we write

$$.7(\sigma_f^{49} N_O^{49} + \sigma_f^{41} N_O^{41} + \sigma_f^{25} N_O^{25})\Theta = 3 \times 10^{20} \quad (3.1.5)$$

An EPITHERMS calculation for the project fuel gave the following values for the cross sections, in barns, averaged over the fuel volume and over the spectrum from zero to ten electron volts.

$$\begin{array}{ll} \sigma_a^{28} = 2.045 \text{ b} & \sigma_f^{25} = 318.9 \text{ b} \\ \sigma_a^{49} = 785.9 \text{ b} & \sigma_f^{49} = 507.7 \text{ b} \\ \sigma_a^{40} = 458.3 \text{ b} & \sigma_f^{41} = 655.4 \text{ b} \end{array}$$

These cross sections take into account the large resonances of U-238, Pu-239, and Pu-240 which occur at low energy. Based on these values, eqn. (3.1.5) gives  $\Theta = 2.2 \times 10^{21} \text{ cm}^{-2}$  or  $2.2 \times 10^{-3} (\text{barns})^{-1}$ .

Based on these values, we evaluate eqns. (3.1.1 - 3.1.4) and obtain

$$\begin{aligned} \frac{d\Delta N^{49}}{\Delta N^{49}} &= .65 \frac{d\sigma_a^{49}}{\sigma_a^{49}} \\ \frac{d\Delta N^{40}}{\Delta N^{40}} &= .8 \frac{d\sigma_a^{49}}{\sigma_a^{49}} - 2.4 \frac{d\sigma_a^{40}}{\sigma_a^{40}} \end{aligned} \quad (3.1.6 \text{ a,b})$$

Consideration of the data obtained from the differential solution experiment and from comparisons of the darkening of several  $\alpha$ -autoradiographs lead to an estimate of the ratio of plutonium activity in hot spots to the activity in the background of about 46 times. Based on the estimate of the activity ratio, which is the same as the density ratio, and taking a nominal 1.5% of the mass of pellets as plutonium oxide, the segregation of the plutonium amounts to 31% of the total plutonium in the pellet.

Taking the distribution of sizes of the hot spots as given by Table 2.4.1, an estimate of the fractional error made in the cross sections gives

$$\frac{\Delta \sigma_a^{49}}{\sigma_a^{49}} = -.073$$

$$\frac{\Delta \sigma_a^{40}}{\sigma_a^{40}} = -.045$$

Equations (3.1.6) give for the errors in the isotopic concentrations

$$\frac{d\Delta N^{49}}{\Delta N^{49}} = -5\%$$

$$\frac{d\Delta N^{40}}{\Delta N^{40}} = 5\%$$

The signs on the errors are such that the amount of Pu-239 remaining after any given exposure would be overestimated and the amount of Pu-240 present would be underestimated compared to measurement of the amounts actually present.

### 3.2 Evaluation of the Fuel

It must be observed that the estimated uncertainties in the effective pellet-average cross sections, due to increased self-shielding in the hot spots in the fuel, are larger than the expected errors arising from uncertainties in the cross sections inferred from the burnup measurements. The fundamental cross section and spectral errors might be expected to yield results good to a few percent, especially if some of the burnup data are used to obtain gross correction factors for uniform fuel.

The errors due to segregation cannot be corrected for in the same way because of the difficulty in determining exactly the size distribution and numbers of hot spots. To obtain a sufficiently large and well analyzed sample of hot spots would seem to require a program beyond the scope of the present work.

If the evaluation of the hot spots could provide a correction factor which would reduce the cross section error by half, the remaining 2% uncertainty of Pu-239 and Pu-240 would be comparable to the accuracy with which the isotopic changes can be determined with the mass-spectrometer.

The errors in the Pu-241 and Pu-242 concentrations have not been analyzed, but it is expected that they would be somewhat larger than the lower isotopes.

Because of the statistical variation of the total Pu content per unit fuel volume, it will be necessary to abandon the fine radial sampling. The samples will have to be restricted to no more than two radial samples of equal volume.

#### IV. CONCLUSIONS

In view of the foregoing analysis, it seems clear that it is necessary to determine more precisely the exact nature of the microsegregation problem and its effect on the post-irradiation plutonium isotope concentrations.

The statistical certainties with which the distribution of sizes and the concentrations of the hot spots are known is at present quite poor. To make a more precise evaluation of these hot spots, it will be necessary to conduct a very extensive investigation by means of more autoradiographs and by mass spectrographic analyses of background and hot spot material extracted by core drilling. This extended fuel analysis activity is beyond the scope of the present Program, but it is necessary in order to establish upper limits for the uncertainties introduced by the microsegregation.

This conclusion may be reinforced by considering that our present knowledge of the fuel is based on only a few samples (statistically speaking); the computed effects of the microsegregation, as stated above, are themselves certain to little better than  $\pm 100\%$ , even assuming the correctness of the concentrations (31% of the total Pu segregated in spots containing 60%  $\text{PuO}_2$ , 40%  $\text{UO}_2$ ) used in the calculations. When allowance is made for the uncertainties in these latter quantities, the calculated consequences of microsegregation are seen to be no more reliable than plus or minus several hundred per cent.

Half of the estimated error in the cross section is introduced by the single 20 and 30 mil spots observed. If an additional 10 pellets are autoradiographed, and if no more 20 or 30 mil spots are observed, the indicated cross section errors would be reduced by 40%, as would the errors in the isotopic concentrations. Thus, the error in the change of the 240 concentration would fall from 5% to 3%.

Similarly, one 20-mil plus one 30-mil hot spot per three pellets represents 15% of the total plutonium. If no more hot spots of this size are found, the expected amount of Pu contained in these large spots would fall to 3.5%. The variation in the total plutonium content from pellet to pellet would then be much less than the present data would indicate.

The EPITHERMØS computation was not completely converged, and information from Hanford indicates a coding error in THERMØS may be responsible. The effect of the error on the above analysis must be evaluated before a completely definite recommendation can be made.

Under these circumstances, it is clear that the fuel should be subjected to a much more thorough examination. It is planned that 10 more pellets will be polished and  $\alpha$ -autoradiographed to improve the knowledge of the size and number of the hot spots. It is also planned that a core drilling procedure will be used to extract small samples from both the hot spots and the background, to obtain a precise mass-spectrographic analysis of the plutonium concentration distribution.

V. FUTURE PLANS

Further examination of the unirradiated fuel pellets, in the manner described in the preceding paragraphs, will be made to improve the knowledge of the distribution of the hot spots and of their composition.

Further work on the EPI THERMOS code is continuing to bring it to a production status.

The fuel irradiation in the VBWR is continuing. At the present rate of exposure, the fuel should reach 5,000 MWD/T sometime in the fourth quarter of 1963.

VI. REPORTS ISSUED SINCE INCEPTION OF THE PROGRAMQUARTERLY REPORTS

| <u>Report Period</u>          | <u>Report Number</u>  |
|-------------------------------|-----------------------|
| April 1 - July 1, 1961        | Letter Report         |
| July 1 - September 30, 1961   | Letter Report         |
| October 1 - December 31, 1961 | GEAP-3902, EURAEC 355 |
| January 1 - March 31, 1962    | GEAP-3916, EURAEC 357 |
| April 1 - June 30, 1962       | GEAP-4051             |
| July 1 - September 30, 1962   | GEAP-4081             |
| October 1 - December 31, 1962 | GEAP-4161             |
| January 1 - March 31, 1963    | GEAP-4216             |

TOPICAL REPORTS

None

**END**