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

**PLUTONIUM SUBCRITICAL
EXPERIMENT PROGRAM
SIXTH QUARTERLY REPORT
OCTOBER 1 - DECEMBER 31, 1965**

J. G. CARVER

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PLUTONIUM SUBCRITICAL EXPERIMENT PROGRAM

Sixth Quarterly Report
October 1 - December 31, 1965

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CONTENTS

	<u>Page</u>
FOREWORD	iv
I. INTRODUCTION	
1.1 Summary of Progress to Date	1
1.2 Principal Investigators	2
1.3 Statement of Problem	2
II. DESCRIPTION OF WORK	
2.1 Pressurized Water System	4
2.2 Fuel Lattice System	5
2.3 Theoretical Studies	6
2.4 Licensing	6
III. PLANS FOR FUTURE WORK	
3.1 January - March 1966	9
3.2 Remainder of Calendar 1966	9
IV. CONCLUSIONS	10
V. BIBLIOGRAPHY	11
VI. APPENDIX	12

FOREWORD

"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.

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PLUTONIUM SUBCRITICAL EXPERIMENT PROGRAM

I. INTRODUCTION

(J. G. Carver)

1.1 Summary of Progress to Date

Orders have been placed for all components of the pressurized system. Shipment of the pressure vessel is expected by January 28, 1966. It is expected that the pressurized system will be completed and the first lattice loaded the first week of March, 1966.

Irradiation chambers have been constructed for the fission rate calibration experiment, and procurement of the necessary solid state diodes is in progress. Two 5 in. by 5 in. NaI well crystals have been received, installed, and checked out. Technique development is complete for the production of the glass beads containing solid solutions of oxides of resonance activation materials; production runs have been made and a high degree of uniformity of each composition has been demonstrated.

All of the 525 lattice fuel rods and 20 special fuel rods have been received. Deviations from specification thus far noted will not handicap the reaction rate measurement tasks of the Program.

Investigations are continuing of the resonance absorption in a fuel region as it is affected by spatial and energy variations of flux in the adjacent moderator.

1.2 Principal Investigators

J. G. Carver, M. A. Robkin, C. R. Porter, F. K. Porter,
E. J. Strain, and R. L. Call of General Electric Company;
and M. P. Lagache of EURATOM.

1.3 Statement of Problem

This Program has two major objectives: First, the experimental determination of basic reactor physics data for plutonium-enriched UO_2 fuel lattices in light water; and, second, the construction of a theoretical model capable of accurately predicting all of the experimental results obtained and, by inference, the operating parameters for a plutonium-enriched, H_2O -moderated power reactor. The reactor physics data required will be obtained by measurements in a subcritical lattice of plutonium-enriched fuel rods. Principal emphasis will be upon measurements related to the fuel cycle.

The need for additional data on plutonium-enriched systems derives partly from the pronounced resonances in plutonium and partly from existing uncertainties in the variation of capture/fission ratios across these resonances, which produces large uncertainties in the fuel cycle physics of plutonium. These resonances are at such energies that a relatively small shift in the slow neutron spectrum can substantially change both the relative absorption rates in the several plutonium isotopes and their capture/fission ratios. These problems were discussed at length in the first Quarterly¹.

Part of the Program effort will be devoted to improving techniques for calculating slow neutron spectra. The most advanced theoretical tools will be employed in the selection of procedures for calculating these spectra, and experimental verification of these calculations will be obtained at a number of points within the experimental fuel lattices by the technique of resonance activation.

Major emphasis will be given to the problem of computing reaction rates once the spectrum is established. To this end, extensive measurements will be made of fission and conversion rates in the several fuel isotopes of the lattice.

Because of the sensitive temperature dependence of these quantities in light water lattices, measurements will be made both at room temperature and at approximately 540°F, within a pressure vessel which is being constructed for use in this Program.

II. DESCRIPTION OF WORK

2.1 Pressurized Water System (R. L. Call)

Purchase orders have been placed for all components of the pressurized system. Some rework was required on the welds in the electrical heater pressure shells but these have now been completed and accepted. No schedule problems are anticipated in the delivery of any other components.

Work on the pressure vessel is essentially complete except for hydrostatic testing which must await delivery of studs and nuts. The first set of nuts was completed and discovered to be oversized and a second set is nearing completion. Shipment of the pressure vessel is now expected by January 28, 1966.

California State inspectors have advised us that strict interpretation of the applicable codes requires that Schedule 80 piping be used throughout the system instead of Schedule 40. This change will introduce no delay in completion of the pressurized system but some added cost for materials and engineering labor has been incurred.

A revised piping and instrumentation diagram has been issued to incorporate the changes that have accumulated. Schematic drawings and wiring diagrams are complete. Bids have been received for installation of the piping and this work will begin as soon as Commission approval is received.

Recent minor delays are thus far being absorbed without delaying the scheduled completion of the pressurized system. All construction and testing is scheduled to be completed and the first lattice loaded during the first week in March 1966.

2.2 Fuel Lattice System

2.2.1 Fuel Rod Procurement

The Plutonium Fuels Development Laboratory autoradiographed the 10 sample pellets provided by the vendor from the dry-blended Lattice Fuel order. These autoradiographs appeared to indicate a higher degree of segregation of the plutonia than had been anticipated in view of the specifications issued for this fuel. The Commission was advised that the expected characterization by the vendor was entirely absent and that, with our present limited knowledge of the fuel, it may not be worthwhile to perform the measurements of flux profile and subcriticality that were planned.

Calculations have been made to determine the extent to which any variation in the effective plutonium content affects flux ratios and multiplications in a subcritical lattice constructed from these fuel rods (variations in effective plutonium content include variations in: [a] self-shielding resulting from plutonia segregation [b] mass of oxide loaded in the finished rods and [c] Pu/U ratio). These calculations, which are described in the Appendix, indicate that a variation of 2% in the effective plutonium content results in a change of 0.2% in the multiplication of a typical lattice and a change of 1% in the ratio of the 1 eV flux to the thermal flux. While it was concluded that a 2 per cent uncertainty in effective plutonium content could be tolerated without seriously compromising the flux profile measurements, the possibility remains that the particle self-shielding effect is equivalent to a Pu enrichment variation of more than 2 per cent.

The entire shipment of 525 lattice fuel rods and 20 special fuel rods has been received from NUMEC. A total of 64 rods has been inspected and all have been transferred to the storage racks. Although a number of deviations from specifications have been observed, nothing has been discovered which would prevent the fuel from being used for the reaction rate measurements which are planned for the Program.

None of the unclad pellets of coprecipitated fuel have been received nor have any autoradiographs of this fuel been inspected. Consequently, nothing is yet known about the homogeneity of the coprecipitated fuel.

Photographs of the fuel rods, handling containers, and storage racks appear in Figures 2.1 and 2.2.

2.2.2 Lattice Parameter Calculations (M. P. Lagache, F. K. Porter)

Some further work was done on critical calculations for 7 lattice spacings at 545°F, and these are almost completed. A recheck has been made of the earlier calculations of the Hanford EBWR critical experiments and no significant errors were disclosed.

2.3 Theoretical Studies (M. A. Robkin)

A solution has been found for the intermediate-resonance theory calculation of the effect of the spatial variation of the flux in the non-absorbing moderator surrounding an infinite slab of non-scattering resonance absorber. The intermediate parameter approach of Goldstein and Sehgal² was applied to defining the intermediate parameter. This approach leads to an anomaly because the volume ratio diverges. A modification to their theory is being explored to define a parameter appropriate to this geometry.

2.4 Licensing (E. J. Strain)

Before the pressurized system can be operated, an appropriate amendment to the NTR Facility License must be issued by the AEC.

Application for this amendment was withheld pending Commission action on a general revision to the NTR Facility License. This general revision was issued October 18. On November 5, Amendment No. 13 to the Facility License Application was filed to request the necessary license amendment for operation of the pressurized subcritical. It is understood that Commission action on this application is more than 50% complete.

FIGURE 2.1 FUEL RODS AND HANDLING BASKET

(a) Storage Racks

(b) Rod Baskets in Position Within Racks

FIGURE 2.2 FUEL ROD HANDLING AND STORAGE EQUIPMENT

III. PLANS FOR FUTURE WORK

3.1 January - March 1966

Installation of the pressurized system piping is expected to start in mid-January. Delivery of the pressure vessel is expected by February 4. Final completion of the pressurized system and loading of the first lattice are expected by the first week in March.

Sample pellets of coprecipitated fuel will be alpha-autoradiographed to confirm their homogeneity.

Work will continue on the investigation of spatial effects in resonance absorption.

Work will resume on calibration of the 5 inch by 5 inch well crystal for the Lanthanum¹⁴⁰ fission product activity from U²³⁵ and the two fissile plutonium isotopes.

3.2 Remainder of Calendar 1966

Measurements will be completed on a total of six configurations, at a rate of approximately six weeks apiece. (One configuration is here defined as one combination of lattice spacing and temperature).

IV. CONCLUSIONS

Completion of the pressurized system by early March now appears to be assured.

The Lattice Fuel appears adequate for purposes of the reaction rate measurements, although some doubt remains whether it will be satisfactory for the flux profile measurements. No data or samples have been received for the Special Fuel.

No serious obstacles have arisen to the granting of the facility license amendment required for operation of the pressurized system on schedule.

Timely receipt of the isotopic fission rate foils from ORNL may soon become the key item in the Program schedule.

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VI. APPENDIX

(M. A. Robkin)

The Lattice Fuel for the Plutonium Subcritical Program shows signs of PuO_2 segregation. The basic effect of a variation in the plutonium segregation from one sample of fuel to another is one of variation in the effective self-shielding of the plutonium, which may be treated as a variation in the effective plutonium concentration. Since a large number of particles of PuO_2 were detected by autoradiography, the variation from rod to rod in the effective concentration is probably small compared to the difference between the effective concentrations in the (dry-blended) Lattice Fuel and the (coprecipitated) Special Fuel. This latter difference is unknown and may possibly amount to several per cent.

Rod-to-rod variations in effective plutonium content include contributions from: (a) self-shielding in segregated plutonia particles, (b) mass of oxide per rod, and (c) the Pu/U ratio. The appearance of the alpha autoradiographs suggests that the first of these does not vary significantly throughout the entire lot of dry-blended fuel. Data supplied by the vendor indicate that variations from sources (b) and (c) amount to less than 2 per cent throughout; so that $\frac{\Delta N^{\text{Pu}}}{N^{\text{Pu}}} \leq 0.02$. Let us consider what effect a small variation,

$\Delta N^{\text{Pu}}/N^{\text{Pu}}$ has on k and the ratio of epithermal to thermal fluxes. For a lattice,

$$k_{\text{eff}} = P\eta fEL \quad (1)$$

where P is the resonance escape probability, η is the ratio of production to destruction in the fuel, f is the thermal utilization, E is the fast fission effect, and L is the net non-leakage probability. Equation (1) may be broken up into a factor which is independent of the plutonium content and a factor which is dependent upon the plutonium.

$$k_{\text{eff}} = (P_u \epsilon_L) (P_{\text{Pu}} \eta f) \quad (2)$$

$$\approx k_u k_{\text{Pu}} \quad (3)$$

We define the following nomenclature:

k_{Pu} = plutonium reactivity contribution

P_{Pu} = plutonium resonance escape probability

η = ratio of production to absorption in fuel

f = thermal utilization

α = ratio of secondaries produced per absorption

g_{Pu} = fraction of neutrons absorbed in the fuel which are absorbed in plutonium

g_u = fraction of neutrons absorbed in the fuel which are absorbed in uranium = $1 - g_{\text{Pu}}$

Σ_a = macroscopic absorption cross section of any component

ϕ = flux in a region of volume V

A_i = absorption in some component = $\Sigma_{a,i} \phi V$, $i = \text{Pu}, \text{U}, \text{M}$ where M refers to the moderator

F = fission production from some component = αA

I = resonance integral

$\xi \Sigma_s$ = slowing down power of homogenized cell

v = volume fraction of the fuel in the cell

Σ_{a1} = cell averaged parasitic capture cross section in the epithermal group

Σ_r = cell averaged removal cross section from the epithermal to the thermal group

$\Sigma_{a2} \equiv$ cell averaged absorption cross section in the thermal group

ρ = ratio of thermal to epithermal flux

$$k_{Pu} = P_{Pu} \eta f \quad (4)$$

$$\frac{1}{k} dk = \frac{1}{k_{Pu}} dk_{Pu}$$

$$= \left[\frac{dP_{Pu}}{P_{Pu} dN^{Pu}} + \frac{d\eta}{\eta dN^{Pu}} + \frac{df}{f dN^{Pu}} \right] dN^{Pu} \quad (5)$$

Now

$$P_{Pu} = \exp \left[- \frac{N^{40} I^{40} v}{\xi \Sigma_s} \right] \quad (6)$$

$$I^{40} = \frac{I_\infty}{\sqrt{1 + \frac{N^{40} I_\infty \gamma}{S + \Sigma}}} \quad (7)$$

$$\eta = \frac{F^{Pu} + F^u}{A^{Pu} + A^u} \quad (8)$$

$$= \frac{\alpha^{Pu} A^{Pu} + \alpha^u A^u}{A^{Pu} + A^u}$$

$$f = \frac{A^{Pu} + A^u}{A^{Pu} + A^u + A^M} \quad (9)$$

For $P_{Pu} \approx 1$, we write

$$\frac{dP_{Pu}}{P_{Pu}} = - \left(1 - P_{Pu} \right) \left(\frac{1}{N^{Pu}} + \frac{1}{I^{40}} \frac{dI^{40}}{dN^{Pu}} \right) dN^{Pu} \quad (10)$$

$$\frac{1}{\eta} d\eta = \frac{(\alpha_{Pu} - \alpha_u) \epsilon_{Pu} \epsilon_u}{\alpha_{Pu} \epsilon_{Pu} + \alpha_u \epsilon_u} \frac{dN^{Pu}}{N^{Pu}} \quad (11)$$

$$\frac{1}{f} df = \epsilon_{Pu} (1 - f) \frac{dN^{Pu}}{N^{Pu}} \quad (12)$$

so that

$$\frac{dK}{K} = \left[- \left(1 - P_{Pu} \right) \left\{ \frac{1 + \frac{1}{2} \frac{NI_{\infty} \gamma}{S + \Sigma_s}}{1 + \frac{NI_{\infty} \gamma}{S + \Sigma_s}} \right\}_{40} + \frac{(\alpha_{Pu} - \alpha_u) \epsilon_{Pu} \epsilon_u}{\alpha_{Pu} \epsilon_{Pu} + \alpha_u \epsilon_u} + \epsilon_{Pu} (1 - f) \right] \frac{dN^{Pu}}{N^{Pu}} \quad (13)$$

Now consider the thermal and epithermal flux in an infinite medium based on homogenizing a unit lattice cell. The source to each group is proportional to the energy width of the group. We denote the epithermal group by Group 1 and the thermal group by Group 2. Then

$$\varphi_1 = \frac{S_1}{\Sigma_{r_1} + \Sigma_{a_1}} \quad (14)$$

$$\varphi_2 = \frac{S_2}{\Sigma_{a_2}} \left(1 + \frac{\Sigma_{r_1}}{\Sigma_{r_1} + \Sigma_{a_1}} \frac{S_1}{S_2} \right) \quad (15)$$

$$\rho = \frac{\Sigma_{a_1} + 2\Sigma_{r_1}}{\Sigma_{a_2}} \quad (16)$$

$$= \frac{\Sigma_{40} + 2\Sigma_{r_1}}{\Sigma_a^{Pu} + \Sigma_a^u + \Sigma_a^M}$$

we find

$$\frac{1}{\rho} d\rho = \left[\frac{\Sigma_{40}}{\Sigma_{40} + 2\Sigma_{r_1}} \left\{ \frac{1 + \frac{1}{2} \frac{N I \gamma}{S + \Sigma_s}}{1 + \frac{N I \gamma}{S + \Sigma_s}} \right\}_{40} - g_{Pu} f \right] \frac{dN^{Pu}}{N^{Pu}} \quad (17)$$

Now, by definition

$$P_{Pu} = \frac{\Sigma_{r_1}}{\Sigma_{40} + \Sigma_{r_1}}$$

so

$$\frac{1}{\rho} \frac{d\rho}{\rho} = \left[\frac{1 - P_{Pu}}{1 + P_{Pu}} \left\{ \frac{1 + \frac{1}{2} \frac{NI\gamma}{S + \Sigma_s}}{1 + \frac{NI\gamma}{S + \Sigma_s}} \right\}_{40} - g_u f \right] \frac{dN^{Pu}}{N^{Pu}} \quad (18)$$

A typical set of parameters is

$$P_{Pu} = 0.9$$

$$\alpha_{Pu} = 1.86$$

$$\left(\frac{NI}{\xi \Sigma_s} \right)_{40} \approx 0.10$$

$$f = 0.8$$

$$\alpha_u = 2.11$$

$$g_{Pu} = 0.8$$

$$\alpha_u - \alpha_{Pu} = .25$$

yielding from Equations (13) and (18)

$$\frac{dK}{K} \approx .1 \frac{dN}{N}$$

(19)

$$\frac{d\rho}{\rho} \approx -0.6 \frac{dN}{N}$$

Thus, for $\frac{dN}{N} \approx .02$,

$$\frac{dK}{K} \approx .2\%$$

$$\frac{d\rho}{\rho} \approx -1.2\%$$

which are both tolerable.

It is concluded that rod-to-rod variations in the Lattice Fuel will not interfere with useful measurements of thermal and resonance flux profiles. The major unknown is the magnitude of the overall reduction in effective Pu concentration resulting from the plutonia segregation. If this reduction is of the order of several per cent, and remains unknown, significant ambiguities will arise in comparing measured and calculated flux ratios and profiles in these lattices.

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