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**PLUTONIUM SUBCRITICAL
EXPERIMENT PROGRAM
THIRD QUARTERLY REPORT
JANUARY 1, - MARCH 31, 1965**

J.G. CARVER

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GENERAL ELECTRIC

ATOMIC POWER EQUIPMENT DEPARTMENT
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PLUTONIUM SUBCRITICAL EXPERIMENT PROGRAM

Third Quarterly Report
January 1 - March 31, 1965

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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.

"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."

PLUTONIUM SUBCRITICAL EXPERIMENT PROGRAM

I. INTRODUCTION

(J. G. Carver)

1.1 Summary of Progress to Date

The contract for fabrication of the lattice fuel rods has been placed by the AEC. Nuclear analysis is continuing for a variety of lattices composed of these rods. An investigation is in progress of the resonance absorption in a lump of fuel as it is affected by spatial and energy variations of flux in the surrounding moderator. Design of the pressurized water system is complete and an order has been placed for fabrication of the pressure vessel.

Plans and foil specifications are complete for measurement of fission and conversion rates, and activation rates in resonance absorbers. Orders have been placed for the counting equipment required for calibration of fission rate measurements. A 5" x 5" NaI well crystal has been set up to serve as a general purpose counter for the program. Technique development is complete for the production of glass beads containing solid solutions of the oxides of resonance activation materials; production runs have been made on most of the compositions, and activation measurements have demonstrated a high degree of uniformity within each composition.

1.2 Principal Investigators

J. G. Carver, M. A. Robkin, C. R. Porter, 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 $540^{\circ}F$, 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)

An order has been placed with the Nooter Corporation, St. Louis, Missouri, for fabrication of the main pressure vessel. The vessel will be built of 304 stainless steel, will have an inside diameter of 36 inches and an overall inside length of 60 inches. Three 3-inch nozzles will be provided in the head to accommodate thimbles for a neutron detector, to be used in conjunction with pulsed neutron measurements. The water will be circulated through two 2-inch nozzles in the wall of the main vessel.

The design is now complete for the physical arrangement of the piping and equipment which constitutes the pressurized system. The canned rotor pump, the strainer, and some of the valves are on order. The design of the electrical heaters is complete and some quotations have been received for their construction.

2.2 The Fuel Lattice

2.2.1 Fuel Procurement (J. G. Carver)

The Atomic Energy Commission has placed a contract with The Nuclear Materials and Equipment Corporation, Apollo, Pennsylvania, for delivery of the lattice fuel the first week of October, 1965. The main load of fuel will consist of 525 rods, containing fuel pellets $1/2$ " in diameter, extending for an active fuel length of two feet. An additional 20 rods will be fabricated of coprecipitated mixed oxides. The composition will be 1.5% PuO_2 in natural UO_2 . Although final allocation has not been made, it appears that the Pu-240 content will be approximately 20 per cent. The fuel will be clad in Zircalloy tubing with a wall thickness of 32 mils.

2.2.2 Lattice Parameter Calculations (M. P. Lagache, M. A. Robkin)

In a December 4, 1964, meeting with J. R. Worden and R. C. Liikala of Hanford, informal agreement was reached on the desirability of a detailed comparison between the Hanford and Vallecitos theoretical descriptions of representative fuel arrays. The lattices selected for this comparison were those of the EBWR criticals at Hanford (reported in HW-83187) and the Hanford 2% - PuAl critical experiments (reported in HW-79054).

Both series of experiments were analyzed using two different energy schemes (a 3-group breakdown, and the 4-group representation in use at Hanford). The few-group cell constants were obtained with a set of three computer codes: a MUFT-type multi-group epithermal computation, a thermal spectrum calculation based on the Wilkins Kernel, and a P3 spherical harmonic expansion. The Hanford code, HFN, was then used to represent the complete cores and compute their multiplications. The computed results for the EBWR criticals were slightly better than those previously obtained (within less than 2% of the measured values at all water-fuel ratios from 1.1/1 to 5/1). The results for the PuAl lattices were also in reasonable agreement with the experimental data (+ 2.5%, - 0). A thorough review of these calculations disclosed no errors in the computer input.

The calculations performed by the Hanford (now Pacific Northwest Laboratories) physics group were uniformly closer to the results of the PuAl critical measurements (within + 1.7%, - 0%) than were the calculations performed here. It is possible that our calculations suffer from some inaccuracy in the treatment of in-cell neutron transport; and the Thermos code is now being applied to these cell calculations.

2.2.3 Driving Flux (M. P. Lagache)

An accurate representation of the axial variation of flux and spectrum in the driven subcritical lattices will require an accurate representation of the NTR core, with the source plate on the top face. Work has been started to

construct a model of this system, based on parameters calculated with Thermos, Epithermos, and P1 or P3 MG. These parameters will be used in diffusion theory calculations made with the HFN or 20 Grand code.

2.3 Reaction Rate Measurement (W. R. Morgan, C. R. Porter)

2.3.1 Neutron Spectrum Indicators

The following elements have been selected as neutron spectrum indicators: Lu, Eu, Ir, Au, Cu, La, W, U, Yb. To date, glasses containing Lu, Eu, Cu, Yb, Na, and W have been made. Uniformity checks have been completed on all but Lu and As. The nonuniformity of 3/4% (one σ) observed is comparable to the precision of the measuring technique. Production of U, In, and Se glasses will proceed as soon as the materials arrive. Production of Au-Ni and Ir-Ni alloys is beginning.

In order to demonstrate the sensitivity of the resonance wire technique to the spectral changes in and around fuel, a mockup fuel element segment was constructed using fourteen 3/8" diameter by 1/2" long pellets, enriched to 4.5 per cent. Nine of these pellets have central holes. Resonance wires were located inside and outside both bare and Cd-covered pellets, as well as in a symmetrical non-fueled section. In the first attempt, fission fragment contamination of the wires made complete counting impossible. The experiment will be repeated in April with provision for fission fragment shielding during irradiation.

A single detection system has been chosen for most of the counting work now contemplated under the Program: fission foils, conversion foils, and resonance activation materials. The system will consist of a 5" by 5" NaI well crystal, amplifiers, and a multi-channel pulse-height analyzer.

2.3.2 Counting Equipment

The well crystal has been received and installed in a specially constructed lead shield. Figure 2.1 shows four steps in the assembly of the shielded detector.

The top of the shielded assembly is visible in Figure 2.4 (d), together with a wand for installing samples to be counted; the sample is placed in a small thimble which is threaded onto the end of the wand, and then inserted in the crystal through the tube which protrudes from the top surface of the lid.

Calibration of the fission rate measurement technique will require the use of solid-state diodes with thin uranium fission foils. A fast amplifier and discriminator system will be required to process the signal pulses from the solid-state diode. This equipment is on order and delivery is expected shortly.

2.4 Theoretical Studies (M. A. Robkin)

The resonance absorption in a lump of fuel is affected by the spatial and energy variations of the flux in the surrounding moderator. Investigation of this effect is continuing. Attention has been focused on the extremely simple configuration of a slab of non-scattering resonance absorber immersed in an infinite non-absorbing moderator with constant isotropic scattering in the laboratory system. The neutron balance for this configuration may be cast in terms of transport theory as follows:

$$\psi(x,u) = 1/2 \int_{-\infty}^{\infty} dx' \Sigma_3(x') \int_{u-\Delta}^u du' \frac{\exp(u'-u)}{1-\alpha} E_1 \left[\int_{x'}^x \Sigma(y) dy \right] \psi(x',u') \quad (2.1)$$

(a)

(b)

(c)

(d)

Figure 2.1. Construction of Shielded NaI Well Counter Assembly

where T is the half thickness of the absorbing slab, and, in this work, $E_n(b)$ is the exponential integral of order n of argument b , $\int_1^{\infty} \frac{e^{-by}}{y^n} dy$. The integral (2.1) contributes to the flux in the slab only from scattering in the moderator.

Equation (2.1) may be averaged over the absorber, and taking advantage of the spatial symmetry of the problem, we find

$$\begin{aligned} \Sigma_a(u) \bar{\varphi}(u) = \frac{\Sigma_M}{2T} \int_T^{\infty} dx' \int_{u-\Delta}^u du' \frac{\exp(u'-u)}{1-\alpha} \left\{ E_2[\Sigma_M(x'-T)] \right. \\ \left. - E_2[\Sigma_M(x'-T) + 2T\Sigma_a(u)] \right\} \psi_M(x', u') \end{aligned} \quad (2.2)$$

where $\bar{\varphi}(u)$ is the spatially averaged flux in the absorber, Σ_M is the moderator scattering cross section, and ψ_M is the moderator flux.

Since the moderator has no absorption, diffusion theory holds very well there. We use a concept from small source theory and require that, at every lethargy, the diffusion solution satisfy the condition that the net current at the surface of the lump be equal to the absorption rate in the lump.

The diffusion equation which holds in the moderator is

$$-D_M \nabla^2 \psi_M(x, u) + \Sigma_M \psi_M(x, u) = \frac{\Sigma_M}{1-\alpha} \int_{u-\Delta}^u du' \exp(u'-u) \psi_M(x, u') \quad (2.3)$$

where $D_M = \frac{1}{3 \Sigma_M}$.

The scattering term has two limiting cases. In the first, the resonance is narrow and the RHS of equation (2.3) is just Σ_M . In the second, the resonance is wide and the RHS is $\Sigma_M \psi_M(x, u)$. We assume that the exact RHS may be represented by a linear combination of wide and narrow resonance forms and write equation (2.3) as

$$-D_M \nabla^2 \psi_M(x, u) + \lambda \Sigma_M \psi_M(x, u) = \lambda \Sigma_M \quad (2.4)$$

where $\lambda = 1$ represents the narrow resonance form, and $\lambda = 0$ represents the wide resonance form. The flux must remain finite; and, applying the boundary condition, the solution to equation (2.4) is

$$\psi_M(x, u) = 1 - \frac{3}{\beta} T \Sigma_a(u) \bar{\varphi}(u) \exp\left[\frac{\beta}{3} (T-x) \Sigma_M\right] \quad (2.5)$$

where $\beta = (3\lambda)^{1/2}$.

Substituting equation (2.5) into equation (2.2), we may perform some of the integrals and we find

$$I(u) = G(u) - F(u) \int_{u-\Delta}^u du' \frac{\exp(u'-u)}{1-\alpha} I(u'), \quad (2.6)$$

where
$$G(u) = \frac{1}{2T} \left\{ \frac{1}{2} - E_3 \left[2T \Sigma_a(u) \right] \right\}, \quad (2.7)$$

$$F(u) = \frac{3}{2\beta^3} \left\{ \beta \left[1 - E_2[2T \Sigma_a(u)] \right] + \left[E_1[2T \Sigma_a(u)] - \exp[2T\beta \Sigma_a(u)] \cdot E_1 \left[2T(1+\beta) \Sigma_a(u) \right] - \ln(1+\beta) \right] \right\}, \quad (2.8)$$

and $I(u) = \Sigma_a(u) \bar{\varphi}(u)$ is the average absorption per unit lethargy in the slab.

Variational solutions are being attempted for the integral equation (2.6).

Equation (2.6) may be solved straight forwardly in the narrow resonance limit. In this case, the integral term is zero, and

$$I(u) = \frac{1}{2T} \left\{ \frac{1}{2} - E_3[2T\Sigma_a(u)] \right\}, \quad (2.9)$$

and, integrating equation (2.9), the resonance integral is found to be

$$I = \frac{8}{3} I_\infty \left(\frac{S}{2\pi\Sigma_a} \right)^{1/2} \quad (2.10)$$

where

$$I_\infty = \frac{\pi\Sigma_a\Gamma}{2E_r}$$

Σ_a = peak resonance absorption cross section

S = reciprocal mean chord length ($1/4T$)

Γ = half-width of resonance

E_r = energy at peak of resonance

The usual NRIA approximation for a lump is

$$I_{\text{NRIA}} = I_\infty \left(\frac{S}{S + \Sigma_a} \right)^{1/2} \quad (2.11)$$

For the case of a slab of uranium metal for which $S \ll \Sigma_a$, the difference between the two results is

$$\frac{\Delta I}{I} = \frac{I - I_{\text{NRIA}}}{I} = 6.4\%, \quad (2.12)$$

independent of slab size so long as $S \ll \Sigma_a$.

The usual NRIA result is based on Wigner's rational approximation for the escape probability from the slab

$$P_e(u) = \frac{S}{S + \Sigma_a(u)} \quad (2.13)$$

Several authors^{2,3} have suggested modifying the rational approximation to

$$P_e(u) = \frac{S}{S + a \Sigma_a(u)} \quad (2.14)$$

I and I_{NRIA} can be made equal by selecting

$$a = \left[1 - \frac{32}{9\pi} \frac{S}{\Sigma_a} \right] \frac{9\pi}{32} \quad (2.15)$$

and for $S \ll \Sigma_a$

$$a = \frac{9\pi}{32} = .88357$$

2.5 Licensing

Before installation of the pressurized system in the NTR cell can begin, a construction permit must be issued by the AEC. Before the system can be pressurized or plutonium-enriched fuel received, an application must be filed and an amendment to the NTR Facility License must be received. Before any of these licensing actions could occur, it was first necessary that the General Electric Company complete its application for a general revision to the present NTR license. This application was filed in March.

Work has begun collecting all available information on possible dispersal of plutonium, and on organizing relevant information on high pressure steam systems. Early next quarter, this information will be assembled into an application for an amendment to the NTR license, which will permit the activities contemplated under this program.

2.6 Schedules

The critical element in the program schedule is the pressure vessel. The present fabrication schedule provides for shipment from St. Louis the last week of November, 1965. It may prove possible to proceed simultaneously with fabrication and with some of the analyses required by Section III of the

ASME Boiler Code; if this can be done, the pressure vessel may be delivered as early as the first week in October. In any event, all other components of the pressurized system will be in place and ready for the pressure vessel to be connected. The fuel fabrication schedule provides for delivery of fuel rods the first week of October, which will be appropriately timed for the earlier delivery date of the pressure vessel.

III. PLANS FOR FUTURE WORK

3.1 April - June 1965

Orders will be placed for all of the remaining auxiliary components of the pressurized system.

All avenues for expediting delivery of the pressure vessel will be vigorously explored.

The electronics equipment for fission rate calibrations will be received, and calibration work with U-235 foils will be started. The response of the 5" x 5" well crystal will be measured for the Lanthanum-140 fission product and for the activated resonance absorber materials.

Applications will be filed with the Commission for appropriate amendment(s) to the NTR facility license to permit installation of the pressurized system and the performance of experiments with plutonium enriched fuels.

IV. CONCLUSIONS

Accurate predictions of multiplications in the subcritical lattices will require further refinement of the analytical techniques now being used in this program.

Oxides of Lu, Eu, Cu, Yb, Na, and W can be dissolved uniformly in glass of an appropriate composition, and the resulting glass can be formed into beads of a convenient size and shape for resonance activation studies.

An integral equation can be set up to describe the absorption in a slab of fuel surrounded by moderator, including the dependence of resonance absorption on spatial and energy variations of flux in the moderator; and variational methods offer considerable promise to obtaining solutions to this equation.

The program schedule is now limited by delivery of the main pressure vessel; reaction rate measurements may begin as late as Jan. 1966 and no earlier than the first of November 1965. It is expected that, by applying intensive effort as soon as the pressurized system is available, measurements can be completed in the first lattice and started in the second lattice by June 1966, and so bring the program within six weeks of its original schedule.

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