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

ELEVENTH QUARTERLY REPORT
OCTOBER 1 - DECEMBER 31, 1963

M.A. ROBKIN

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GENERAL ELECTRIC
ATOMIC POWER EQUIPMENT DEPARTMENT
SAN JOSE, CALIFORNIA

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

Eleventh Quarterly Report

October 1 - December 31, 1963

by

M. A. Robkin

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APPROVED:

M. A. Robkin
Project Scientist

J. G. Carver, Manager
Fuels and Irradiations Physics

T. M. Snyder, Manager
Physics Sub-Section

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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

Forty-six fuel pellet faces have been autoradiographed. These faces have been prepared from twenty-three pellets by making an exposure before and after the removal of an additional ten mils of fuel.

A substantial number of large "hot spots" continue to appear. The largest spot so far observed was 44 mils long, 20 mils wide, and of the order of 20 mils thick. This spot had a PuO_2 concentration which varied from 70% on the periphery to 100% at the center.

There is some evidence that the segregated regions are elongated with their long axes perpendicular to the direction of the pressing of the green pellet. Determination of the size and concentration distribution is continuing.

The EPITHERMØS code now seems to be operating correctly. A test problem for a typical water lattice converged in eleven iterations. The computation of the spectrum for a pure water medium gave results which agreed very well with the expected 1/E spectrum.

At the end of the quarter, the program fuel element had received a cumulative total of 4449 MWD/T exposure. This total 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 5306 MWD/T.

Three sets of flux wires were successfully irradiated at three thimble locations in the project fuel element. Counting is in progress and the data will be reduced in the next quarter.

The program fuel element was removed from the VBWR during the November shutdown at the end of run 165 after a cumulative exposure of about 5000 MWD/T. Fuel pin F was removed for analysis, and pin U was moved over into its place. An unirradiated fuel pin, designated M, was inserted in the position vacated by U. The reassembled element was returned and reinserted in the VBWR.

The VBWR discontinued operations in December, 1963. The project fuel element was removed from the reactor and placed in storage in the Fuel Storage Building.

1.3 Principal Investigators

M. A. Robkin and C. R. Porter of the 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 exposure of 746 MWD/T during the quarter, bringing the total exposure to 4449 MWD/T. This total is as logged by VBWR operating personnel. Applying the same correction factor, between logged exposure and Ce-Cs analysis of the first fuel sample, gives a corrected exposure of 5306 MWD/T.

The fuel irradiation was terminated with the discontinuation of operation of the VBWR. The project fuel element was removed from the reactor and placed in storage in the Fuel Storage Building.

2.2 Flux Wire Experiment

Four sets of flux wires were irradiated in thimbles inserted in the program element at the November shutdown. One set was lost on removal through the valve and three sets were successfully retrieved and transferred to Physics for counting. The positions in the element from which wires were retrieved are those designated Nos. 1, 2, and 3, in the Northwest, Northeast, and Southeast thimble positions, respectively. These positions are shown on Page 11 of Section IV in the Sixth Quarterly Report, GEAP-4081. The set of activants used in these flux wires, and the neutron energies to which they respond, are listed on Page 31 of Subsection 4.2.1.

Counting of the activated wires is in progress, and the reduction of the data is expected to be completed in the next quarter.

2.3 Autoradiography

Examination of the unirradiated project fuel continued with the preparation of a sequence of polished faces. Twenty-three pellets were used to produce two sets of autoradiographs, the "A" sequence and the "B" sequence. Every member of the "A" sequence has a corresponding member of the "B" sequence which was prepared by removing an additional 10 mils from the face used to make the "A" autoradiograph.

The 10-mil increment was used to determine the persistence of features found on the "A" face, so that some idea of the "thickness" of the hot spots could be obtained. Of the 23 "A" faces examined, only two showed a spot which persisted after 10 mils additional grinding, which suggests that the segregated

PuO_2 occurs as flattened shapes. Further evidence for this conclusion is provided by some autoradiographs of sections cut parallel to the axes of the pellets, as prepared by J. B. Burnham of Hanford. In these autoradiographs, the flattening is quite apparent. The PuO_2 regions appear to be oriented with their long dimension perpendicular to the direction of pressing of the green pellets from the mixed powders (i.e., with their long dimension normal to the axis of the pellet).

Large "hot spots" continued to appear. In the 46 faces, 13 spots larger than 15 mils were observed, with the largest being an irregular triangular shape with a 20-mil base and 44-mil altitude. These large spots were all of high PuO_2 content, running well above 50% for most, with the largest at nearly pure (100%) PuO_2.

Estimates of size, shape, and concentration now indicate that about 10% of the total PuO_2 is segregated as large, high concentration "hot spots", with an additional small amount segregated as small, lower concentration spots. The total segregated amount is estimated on the basis of all of the autoradiographs to be in the neighborhood of 10% (to within a factor of two).

2.4 Fuel Sampling

The Program fuel element was removed from the VBWR during the November shutdown at the end of run 165 after a cumulative exposure of about 5000 MWD/T. Fuel pin F was removed for analysis and pin U was moved over into its place. An unirradiated fuel pin, designated M, was inserted into the position vacated by U. The reassembled element was returned and reinserted into the VBWR.

The above fuel manipulation corresponds to the first step in an irradiation sequence designed to utilize two homogeneous coprecipitated UO_2-1.5% PuO_2 fuel pins which were to have been fabricated in January, 1964. With the shutdown of the VBWR, the proposal to fabricate two coprecipitated fuel pins has been abandoned.

The fuel element was examined visually in the Radioactive Materials Laboratory during its dismantling and reassembly. Figure 2.4.1 shows four views of the element prior to disassembly. Some surface discoloration is evident, but no signs of significant corrosion, damage, or unusual conditions are apparent.

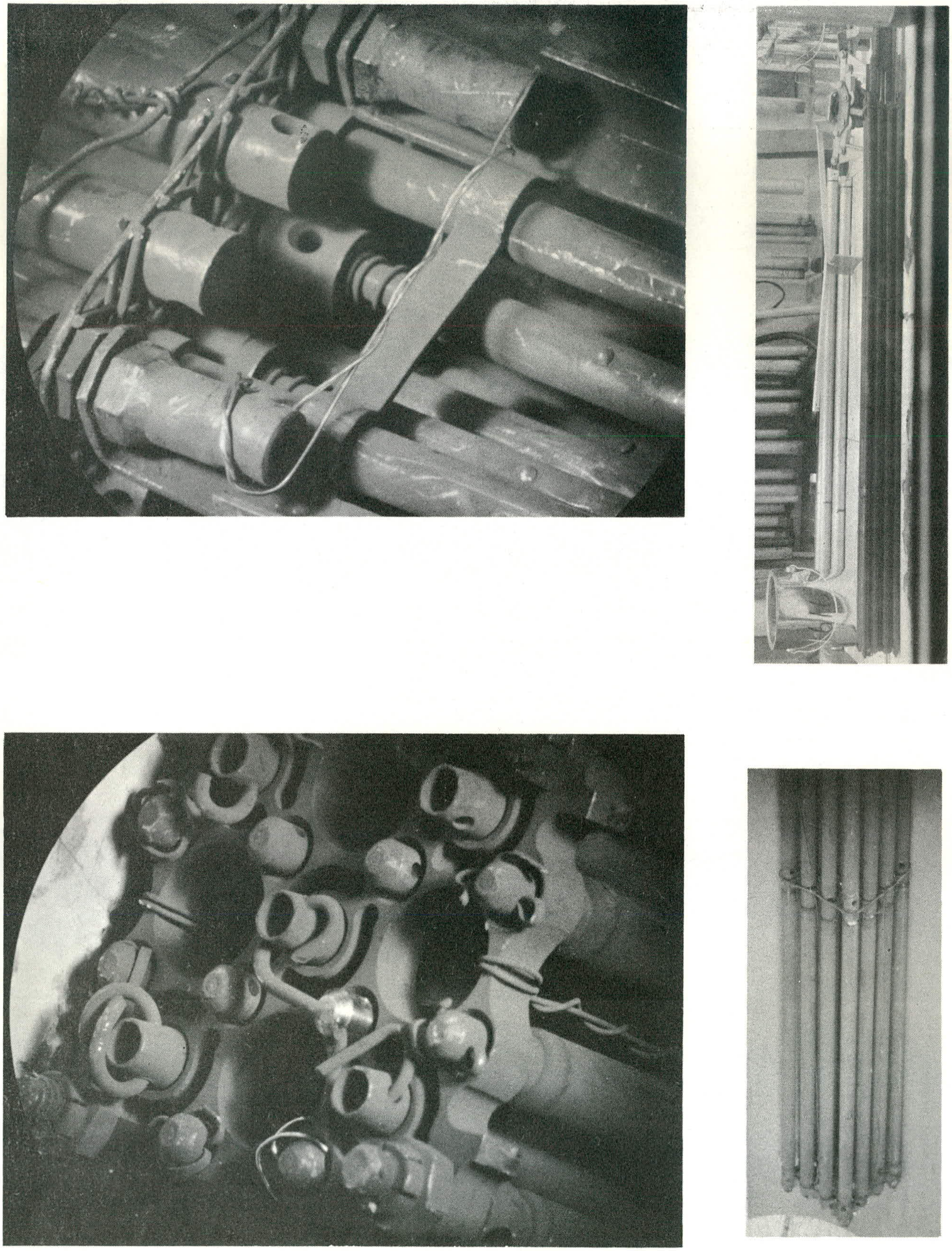

Figure 2.4.1 Some Views of the Program Fuel Element Prior to Removal of the 5000 MWD/T Pin

III. DESCRIPTION OF WORK - THEORETICAL

3.1 EPITHERMØS

The difficulty in the convergence of the epithermal part of the code has been eliminated. The failure of the calculation to converge was traced to the use of the value of the initial flux guess at the center of the geometry and at the tenth energy point. For those cases where this flux value is not the maximum (as assumed by the code), it takes much longer for the calculation to converge to an error which is a given fraction of this reference value.

The code was corrected by requiring that it search through the initial flux guess to find the largest value. The number of iterations required to satisfy the convergence requirement fell from over 100 to 11 for a particular typical case tried.

The library cross section for Pu-240 was adjusted to give the resonance integral cross section for infinite dilution corresponding to a 1/E spectrum between .785 and 10 ev. The value of 8300 barns was chosen as the best value for the resonance integral.

For the case of pure water, after 11 iterations, the first three energy moments and the dilute Pu-240 resonance integral were as follows, where the theoretical values correspond to a 1/E spectrum.

TABLE 3.1.1

	$\overline{V}$	$\overline{V^2}$	$\overline{V^3}$	I_{240}	$\Delta\overline{V}$	$\Delta\overline{V^2}$	$\Delta\overline{V^3}$	ΔI_{240}
	relative units				(%)			
Theory	9.858	110.7	1409	8300				
EPITHERMØS	9.880	111.4	1418	8315	+.22%	+.63%	+.64%	+.18%

IV. CONCLUSIONS

As additional faces of the unirradiated project fuel are examined, the same extensive microsegregation of PuO_2 into large volumes continues to be observed.

Insofar as these large spots (10 mils or larger) are concerned, a 10-mil difference between faces for 46 faces represents the examination of a volume about equal to that of one pellet. This reasoning is based on assigning a typical thickness of 10 mils to each large hot spot; and, since practically every face has at least one hot spot as large as, or larger than, 10 mils, about 1/2 inch of pellet is examined for large spots.

Sufficient statistics have accumulated to consider that, to within a factor of two, about 10% of the plutonium may be considered to be segregated as high concentration PuO_2. On the basis of the sizes observed, the effect of the "hot spots" on the reaction rates in the fuel will be re-evaluated.

For determining the burnup and isotopic composition of the fuel in the presence of the extensive segregation, a sampling procedure is needed which averages out the segregated plutonium in a large sample volume. It is proposed to obtain samples which constitute about half a pellet each by use of a quarter-inch drill to core out the centers of each of three pellets. Six samples will be obtained, of which three will be "inner" half pellets and three will be "outer" half pellets.

Since the sampling procedure used for the 1860 MWD/T fuel pin was based on radial grinding of small annular volumes, the statistical uncertainty in the plutonium content and isotopic composition, due to the microsegregation, largely invalidates the subsequent assays of the samples so obtained.

By performing assays on new samples obtained from the stored 1860 MWD/T pin, by the large-volume drilling described above, it will be possible to use both exposure points (1860 and 5000 MWD/T) to verify the procedure developed to correct the isotopic evolution for the effect of microsegregation.

The EPITHERMØS code now seems to be working very well for water lattices, and computes a Pu-240 resonance cross section which is in good agreement with the value reported in the literature (8300 barns). Comparison of the computed relative reaction rates for the resonance activants with the measured values will serve to confirm the reliability of the code.

V. FUTURE PLANS

The reduction of the data obtained in the flux wire irradiation experiment will be completed.

The large-volume sampling procedure will be carried out on three central pellets of the pin exposed to 5000 MWD/T, and on three surviving pellets closest to the center of the plutonium region in the pin which was exposed to 1860 MWD/T. The samples will be analyzed for burnup and for the plutonium and uranium isotopic composition.

A theoretical evaluation of the effect of segregation on reaction rates will be made and the resulting computed isotopic composition will be compared to the experimental analysis.

A comparison will also be made between the results of the flux wire activation experiment and the relative reaction rates computed by EPITHERMØS.

VI. REPORTS ISSUED SINCE INCEPTION OF THE PROGRAM

QUARTERLY REPORTS

Report Period	
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
April 1 - June 30, 1963	GEAP-4303
July 1 - September 30, 1963	GEAP-4385

TOPICAL REPORTS

None