AUG 19 1965

GEAP-4912
JULY 1, 1965

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

1442

MASTER

**PLUTONIUM SUBCRITICAL
EXPERIMENT PROGRAM
FOURTH QUARTERLY REPORT
APRIL 1 - JUNE 30, 1965**

J. G. CARVER
M. A. ROBKin

APPROVED FOR PUBLIC RELEASE

U. S. ATOMIC ENERGY COMMISSION
CONTRACT AT(04-3)-189
PROJECT AGREEMENT 38

VALLECITOS ATOMIC LABORATORY

GENERAL ELECTRIC

ATOMIC POWER EQUIPMENT DEPARTMENT
SAN JOSE, CALIFORNIA

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

EURAEC
GEAP-4912
Joint U.S.-EURATOM Research
and Development Program

July 1, 1965

PLUTONIUM SUBCRITICAL EXPERIMENT PROGRAM

Fourth Quarterly Report
April 1 - June 30, 1965

J. G. Carver

and

M. A. Robkin

APPROVED:

JG Carver *MARobkin* *G. T. Petersen* *Yun Wall*
J. G. Carver, Mgr. G. T. Petersen, Acting Mgr.
Fuels & Irradiations Physics Physics Sub-Section
(Project Scientist)

Prepared under Contract AT(04-3)-189, P.A. No. 38

Printed in U.S.A. Available from the
Clearing House for Federal Scientific and Technical Information
National Bureau of Standards, U. S. Department of Commerce
Springfield, Virginia

Price: \$2.00 per copy.

VALLECITOS ATOMIC LABORATORY

2454 TIO-2
80 DAC 6/65

GENERAL ELECTRIC

ATOMIC POWER EQUIPMENT DEPARTMENT
SAN JOSE, CALIFORNIA

LEGAL NOTICE

THIS DOCUMENT WAS PREPARED UNDER THE SPONSORSHIP OF THE ATOMIC ENERGY COMMISSION PURSUANT TO THE JOINT RESEARCH AND DEVELOPMENT PROGRAM ESTABLISHED BY THE AGREEMENT FOR COOPERATION SIGNED NOVEMBER 8, 1958, BETWEEN THE GOVERNMENT OF THE UNITED STATES OF AMERICA AND THE EUROPEAN ATOMIC ENERGY COMMUNITY (EURATOM). NEITHER THE UNITED STATES, THE U.S. ATOMIC ENERGY COMMISSION, THE EUROPEAN ATOMIC ENERGY COMMUNITY, THE EURATOM COMMISSION, NOR ANY PERSON ACTING ON BEHALF OF EITHER COMMISSION:

- A. MAKES ANY WARRANTY OR REPRESENTATION, EXPRESS OR IMPLIED, WITH RESPECT TO THE ACCURACY, COMPLETENESS, OR USEFULNESS OF THE INFORMATION CONTAINED IN THIS DOCUMENT, OR THAT THE USE OF ANY INFORMATION, APPARATUS, METHOD, OR PROCESS DISCLOSED IN THIS DOCUMENT MAY NOT INFRINGE PRIVATELY OWNED RIGHTS; OR
- B. ASSUMES ANY LIABILITIES WITH RESPECT TO THE USE OF, OR FOR DAMAGES RESULTING FROM THE USE OF ANY INFORMATION, APPARATUS, METHOD OR PROCESS DISCLOSED IN THIS DOCUMENT.

AS USED IN THE ABOVE, "PERSON ACTING ON BEHALF OF EITHER COMMISSION" INCLUDES ANY EMPLOYEE OR CONTRACTOR OF EITHER COMMISSION OR EMPLOYEE OF SUCH CONTRACTOR TO THE EXTENT THAT SUCH EMPLOYEE OR CONTRACTOR OR EMPLOYEE OF SUCH CONTRACTOR PREPARES, HANDLES, DISSEMINATES, OR PROVIDES ACCESS TO, ANY INFORMATION PURSUANT TO HIS EMPLOYMENT OR CONTRACT WITH EITHER COMMISSION OR HIS EMPLOYMENT WITH SUCH CONTRACTOR.

To expedite dissemination of technical information to those persons requiring it, this report has been issued without the administrative review necessary to protect the interest of the U.S. Government, the European Atomic Energy Community (Euratom) and the contractor. The recipient of this report agrees that it is passed on to him in confidence, that he will maintain that confidence and make no disclosures or publication of the contents, that would be detrimental to the interests of the U.S. Government, the European Atomic Energy Community (Euratom) and the contractor.

CONTENTS

	<u>Page</u>
FOREWORD	iv
I. INTRODUCTION	
1.1 Summary of Progress to Date	1
1.2 Principal Investigators	1
1.3 Statement of Problem	2
II. DESCRIPTION OF WORK	
2.1 Pressurized Water System	4
2.2 Fuel Lattice	4
2.3 Reaction Rate Measurements	10
2.4 Theoretical Studies	13
2.5 Licensing	14
2.6 Schedules	15
III. PLANS FOR FUTURE WORK	
3.1 July - September 1965	16
IV. CONCLUSIONS	17
V. BIBLIOGRAPHY	18

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. A purchase order has been placed for fabrication of the pressure vessel, and delivery has been promised the end of November, 1965. Most of the major components of the pressurized system have been specified and purchase orders are in process. Investigation is continuing of the resonance absorption in a lump of fuel as it is affected by spatial and energy variations of flux in the surrounding moderator.

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

1.2 Principal Investigators

J. G. Carver, M. A. Robkin, C. R. Porter, F. K. 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°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)

Design drawings, and fabrication procedures for the main pressure vessel (Figure 2.1), have been submitted by the vendor, Nooter Corporation of St. Louis, Missouri. These have been reviewed and found generally acceptable. It will be necessary to revise the proposed composition for the main bolts of the vessel. Nooter Corporation has advised that their supplier has made a significant error in the composition of the forging to be used for the flanges of the vessel; an evaluation of the composition actually achieved leads to the conclusion that the flange stock must be refabricated.

Orders have been placed for the electrical heaters, the positive displacement pump, and some of the instrumentation for the pressurized system. (Figures 2.2, 2.3, 2.4, and 2.5).

2.2 Fuel Lattice2.2.1 Fuel Procurement (J. G. Carver)

The vendor for the lattice fuel, Nuclear Materials and Equipment Corporation, Apollo, Pennsylvania, has submitted quality control and production plans for approval by the Atomic Energy Commission. Comments and recommendations concerning these plans have been provided to the AEC.

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

The analysis of the EBWR and the 2% PuAl Hanford critical experiments was completed, using a somewhat more sophisticated model than the one previously used. So far, cell parameters have been computed by a Wilkins-P3 code, and, above thermal, by a code similar to Muft. These parameters, used in a diffusion theory calculation, led to an

FIGURE 2.1 P₀ SUBCRITICAL EXPERIMENT PRESSURE VESSEL

FIGURE 2.2 Pu SUBCRITICAL EXPERIMENT

(In-cell equipment arrangement - plan view)

FIGURE 2.3 Pu SUBCRITICAL EXPERIMENT

(In-cell equipment arrangement - looking east)

FIGURE 2.4 P_u SUBCRITICAL EXPERIMENT

(In-cell equipment arrangement - looking south)

FIGURE 2.5 Pu SUBCRITICAL EXPERIMENT

overestimation of the PuAl k_{eff} with a maximum error of 2.6% and an underestimation of the EBWR k_{eff} by as much as 2% (for the tightest lattice investigated).

The new calculational technique relies chiefly on a Muft-type calculation above thermal, but uses Epithermos to compute the Pu-240 effective resonance integral and Thermos to represent the thermal region. This technique led to a smaller disagreement in the PuAl lattices (maximum error 1.7%) but led to a somewhat greater disagreement in the EBWR criticals (3%). It is believed that these last calculations should be thoroughly reviewed before the above results can be accepted as final; they include a number of hand-computed corrections which had to be introduced in the course of developing this new procedure.

A review of the above calculations has been delayed in order that the calculations of the P. A. 38 proposed lattices could be started without further delay in order to supply, on time, much needed information on k_{eff} , spectra, flux levels, etc. As soon as all necessary data has been secured (in first approximation), work on the calculation model itself will be resumed.

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

2.3.1 Neutron Spectrum Indicators

The complete set of resonance absorption materials, as listed in the following table, has now been produced.

<u>Material</u>	<u>Carrier</u>	<u>Neutron Resonance or Threshold Energy</u>
Lu ¹⁷⁶	Glass	0.143 eV
Eu ¹⁵¹	Glass	0.461 eV
Yb ¹⁶⁸	Glass	0.597 eV
Ir ¹⁹¹	Ni	0.654 eV

Table - Con't.

<u>Material</u>	<u>Carrier</u>	<u>Neutron Resonance or Threshold Energy</u>
Ir ¹⁹³	Ni	1.303 eV
In ¹¹⁵	Glass	1.457 eV
Au ¹⁹⁷	Ni	4.906 eV
W ¹⁸⁶	Glass	18.8 eV
Se ⁷⁴	Glass	27.0 eV
As ⁷⁵	Glass	47.0 eV
La ¹⁴⁰	Glass	73.5 eV
Na ²³	Glass	2.85 keV
U ²³⁸	Glass	5 to 1000 eV
Ni ⁵⁸ in carrier	Ni	3 MeV
Al ²⁷ in carrier	Glass	7.5 MeV
Cu ⁶³	Glass/Cu	1/v

The uniformity of these materials has been checked by activation in an identical flux; the variation between indicated concentrations within glass beads (or nickel samples) has been less than 3/4% for all but Se, which shows a non-uniformity of approximately 1 1/2% (1 σ).

The resonance materials, counting equipment, techniques and procedures were given a rigorous test when 380 resonance beads were activated during run #3716 of the Mixed Spectrum Critical Assembly, and subsequently counted. Preliminary data obtained from these measurements appear to exhibit an encouraging degree of self-consistency.

In order to evaluate the sensitivity of the resonance activation technique to spectrum changes from the outside to the inside of a fuel rod, a mockup fuel element segment was constructed from fourteen 3/8 inch diameter by 1/2 inch long pellets of 4.5% enriched UO₂.

Resonance wires were located inside central holes of some of the pellets as well as on the outside, both bare and Cd covered; and the assembly was irradiated in the Nuclear Test Reactor. For most of the resonance materials, the ratio of resonance absorber activity to copper activity changed by only a few percent between the interior and outside of the fuel rod. These results are reasonable for the bare materials in view of the diameter, enrichment, and composition of the fuel. Essentially similar results were obtained, however, for the cadmium covered materials, for which rough calculations had indicated that larger differences should be observed. It should be emphasized that significant differences in activation ratios should be observable from the inside to the outside of the project fuel, because of its larger diameter and plutonium enrichment.

2.3.2 Counting Equipment

The 5 inch by 5 inch NaI well crystal has been located in a contoured lead shield providing a minimum of 8 inches of lead around the detector. Background studies still going on have led to the addition of Cd and Cu liners between the lead and the crystal to degrade lead x-rays. Enough paraffin has been ordered to shield the assembly from NTR and MSSCA-produced fast neutrons. The detector shows unusually good resolution ($< 8\%$ FWHM for Cs^{137}) for a crystal of that size with a central well.

2.3.3 Fission Rate Calibration Measurements

Nanosecond electronics and solid-state-diode fission fragment detectors have been received for an experiment designed to calibrate the 5 inch by 5 inch NaI well crystal for fission rate foils. Thin, unclad foils of U^{235} , Pu^{239} , and Pu^{241} for this experiment have been received. Work is underway on the construction of irradiation capsules for the foil-detector sandwiches. Procedures are being drafted for the safe handling of the foils and an application is being written

for an appropriate facility license amendment. In the meantime, the calibration program will be started, using the U^{235} .

2.4 Theoretical Studies (M. A. Robkin)

The study of the configuration of a slab of non-scattering resonance absorber immersed in an infinite non-absorbing moderator, with constant isotropic scattering in the laboratory system, continued with the consideration of the wide resonance limit⁽²⁾.

In this case, one begins by finding the solution to the diffusion equation in the moderator with the linear combination of wide and narrow resonances.

$$-D\nabla^2\varphi + \Sigma_s\varphi = \lambda\Sigma_s + (1 - \lambda)\Sigma_s\varphi \quad (1)$$

which yields the solution

$$\varphi = 1 - \frac{3T \Sigma_a(u) \psi(u)}{\sqrt{3\lambda}} e^{\sqrt{3\lambda} \Sigma_s(T-z)} \quad (2)$$

Where T is the slab half-thickness, $\Sigma_a(u)$ is the macroscopic absorption cross section as a function of lethargy, $\psi(u)$ is the volume-averaged flux in the slab, Σ_s is the macroscopic moderator scattering cross section, λ is the linear combination variable, and z is the spatial variable measured from the center of the slab.

An integral transport equation can be written for the spatially averaged lethargy dependent absorption in the slab which may be reduced to

$$A(u, \lambda) = A(u, 1) - F(u, \lambda) \int_{u-\Delta}^u du' \frac{e^{u'-u}}{1-\alpha} A(u', \lambda) \quad (3)$$

Where $A(u, \lambda) = \Sigma_a(u) \psi(u, \lambda)$, the constants, Δ , α of the scattering integral refer to the atomic weight of the moderator and $A(u, \lambda)$ is the effective spatially averaged absorption rate in the slab, and $A(u, 1)$ is the NR result.

In the WR limit, $\lambda \rightarrow 0$ and (3) may be written

$$A(u, 0) = \lim_{\lambda \rightarrow 0} \frac{A(u, 1)}{1 + F(u, \lambda)} \quad (4)$$

Now, it can be shown that

$$\lim_{\lambda \rightarrow 0} F(u, \lambda) = \frac{3}{2T\sqrt{3\lambda}} f(u) \quad (5)$$

where $f(u)$ is a well behaved function. Thus

$$\lim_{\lambda \rightarrow 0} \frac{A(u, 1)}{1 + F(u, \lambda)} = 0 \quad (6)$$

and $A(u, 0) \rightarrow 0$.

This is a physically reasonable result for the WR limit since there is finite absorption in the slab and no slowing down in the moderator.

Work is continuing in evaluating the linear variable λ for the general case when the resonance is neither wide or narrow with respect to a moderator scattering.

2.5 Licensing

Before the pressurized system can be installed or operated, or plutonium-enriched fuel received, appropriate construction permits and amendments to the NTR Facility License must be issued by the AEC. Work is largely complete on collecting all available information on a possible displacement of plutonium, and has begun on analysis of the pressure transient which might follow an accidental release of

steam to the reactor cell. Submittal of this information to the AEC, with an application for these licensing actions should await completion of Commission action on a general revision to the present NTR license; negotiations are in progress on this general revision.

2.6 Schedules

The critical element in the program schedule is still the pressure vessel. The present fabrication schedule provides for shipment from St. Louis the last week of November, 1965. No serious prospects remain for significantly advancing this date.

III. PLANS FOR FUTURE WORK

3.1 July - September 1965

Orders will be placed for all of the instrumentation for the pressurized system. Essentially all components of the system should be received by the end of that quarter.

Work will begin 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.

An irradiation of resonance beads will be performed at the Stanford Pool Reactor to establish the response of the beads to a Maxwellian spectrum.

The general revision to the NTR license is expected to be issued before September, 1965. Following this revision, the ORNL source plate will be received and measurements will be made of its criticality worth on the top face of the NTR.

The calculations on the interaction between the fission plate and the NTR core are in progress. The codes which are currently contemplated for usage for these calculations are being brought into operating condition. As soon as these codes are ready, the interaction calculations will be done.

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

IV. CONCLUSIONS

All of the resonance activation materials of current interest can be dissolved uniformly in a glass of appropriate composition, and beads formed from this glass have been found useful and appropriate for resonance activation measurements.

The analysis of the EBWR and Pu-Al criticals has been completed. The agreement with the measured reactivities is still no better than about 1 1/2% high in the best cases and about 3% low in the worse case. Pending review of the calculational procedures with Hanford personnel, no further work is contemplated in the coming quarter. However, the agreement, insofar as subcritical reactivity is concerned, is adequate for the preliminary design of the proposed first lattice which will be in the region of metal-to-water ratio of maximum reactivity where the curve of reactivity versus metal-to-water ratio is changing the most slowly.

The calculation of the spatial effect in resonance absorption is still in progress.

The program schedule is now limited by delivery of the main pressure vessel; reaction rate measurements may begin as late as January 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 being the program within six weeks of its original schedule.

V. BIBLIOGRAPHY

1. GEAP-4730 (EURAECE-1234) Plutonium Subcritical Experiment Program - Program Summary and First Quarterly Report, July 1 - September 30, 1964, J. G. Carver, October 1964.
2. GEAP-4845 (EURAECE 1374) Plutonium Subcritical Experiment Program - Third Quarterly Report, January 1 - March 31, 1965, J. G. Carver, April 1, 1965.

10484

DISTRIBUTION OF QUARTERLY, TOPICAL,
AND FINAL REPORTS
FOR CONTRACT AT(04-3)-189, P. A. 38

No. of Copies

USAEC
San Francisco Operations Office
2111 Bancroft Way
Berkeley 4, California
ATTN: W. H. Brummett, Jr.

(25)

USAEC
Division of Reactor Development
Washington 25, D. C.
ATTN: Assistant Director

(1)

USAEC
Division of Reactor Development
Washington, D. C.
ATTN: S. Strauch, Reactor Physics Branch

(1)

USAEC
Chicago Operations Office
9700 South Cass Avenue
Argonne, Illinois
ATTN: Director, Reactor Programs Division

(1)

Argonne National Laboratory
9700 South Cass Avenue
Argonne, Illinois
ATTN: A. Shuck

(1)

Babcock and Wilcox Company
Atomic Energy Division
P. O. Box 1260
Lynchburg, Virginia

(1)

Oak Ridge National Laboratory
P. O. Box X
Oak Ridge, Tennessee
ATTN: William D. Manly

(1)

Battelle Memorial Institute
Pacific Northwest Laboratory
P. O. Box 999
Richland, Washington
ATTN: O. Wick

(1)

AT(04-3)-189, P. A. 38

No. of Copies

Combustion Engineering, Inc.
 Nuclear Division
 Prospect Hill Road
 Windsor, Connecticut
 ATTN: Dr. R. H. Gale

(1)

Westinghouse Electric Corporation
 Atomic Power Department
 Pittsburgh 30, Pennsylvania
 ATTN: Dr. R. J. Creagan

(1)

Allis-Chalmers Manufacturing Company
 Nuclear Energy Division
 Milwaukee 1, Wisconsin
 ATTN: C. B. Graham

(1)

United Nuclear Corporation
 Joint U.S.-EURATOM Program
 "Out-of-Pile Evaluation of Mixed
 Uranium-Plutonium Carbides"
 5 New Street
 White Plains, New York

(1)

Commissariat a l'Energie Atomique
 Joint U.S.-EURATOM Program
 "Study of Recycling of Plutonium"
 69 rue de Varenne
 Paris 7, France

(1)

Centre d'Etude de l'Energie Nucleaire
 Joint U.S.-EURATOM Program
 "Use of Plutonium as Fuel in Nuclear Reactors"
 Belgo Nucleaire - CEN
 35 rue des Colonies
 Brussels 1, Belgium

(1)

Nuclear Materials and Equipment Corporation
 P. O. Box 266
 Apollo, Pennsylvania
 ATTN: K. Puechl

(1)

Battelle Memorial Institute
 505 King Avenue
 Columbus 1, Ohio
 ATTN: R. F. Dickerson

(1)

AT(04-3)-189, P. A. 38

No. of Copies

Carborundum Company
P. O. Box 337
Niagara Falls, New York
ATTN: K. Taylor

(1)

Los Alamos Scientific Laboratory
P. O. Box 1663
Los Alamos, New Mexico
ATTN: R. D. Baker

Battelle Memorial Institute
Pacific Northwest Laboratory
P. O. Box 999
Richland, Washington
ATTN: F. G. Dawson

(1)