MASTER

SEP

NUCLEAR MATERIALS AND EQUIPMENT CORPORATION
APOLLO, PENNSYLVANIA

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission

A. Makes any warranty or representation expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report or that the use of any information, apparatus, method, or process disclosed in this report 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 report

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor

NUMEC P-34

September 5, 1961

Mr. Morris Goldberg, Director
Contracts Division
U. S. Atomic Energy Commission
New York Operations Office
376 Hudson Street
New York 14, New York

Attention: Mr. Seymour Zirin, Contracts Administrator

Subject: CONTRACT AT(30-1)-2389
"DEVELOPMENT OF PLUTONIUM BEARING FUEL MATERIALS"
MONTHLY PROGRESS LETTER FOR MONTH OF AUGUST 1961

Gentlemen:

NUMEC herewith transmits the Monthly Progress Letter for August 1961 covering work performed under Contract AT(30-1)-2389.

PROJECT AND FACILITY ADMINISTRATION

A shipment of 1.5 kg of plutonium nitrate was received on August 26, 1961. Processing of this material was started immediately in order to provide fuel for the initial in-pile rabbit tests. Pure PuO_2 and coprecipitated PuO-UO_2 are being prepared for these tests in order to evaluate the differences in thermal performance between mechanically-mixed oxide fuels and coprecipitated material.

The outfitting of the autoclave and NaK canning glove boxes is nearing completion. These will be ready for commitment to plutonium as soon as required for the preparation of the irradiation samples.

Near the end of this period a molybdenum-wound sintering furnace failed. This furnace is now being disassembled to find the cause of failure.

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.

PREPARATION AND CHARACTERIZATION OF FUEL MATERIALS

Preparation of a 400 gm lot of UO_2 -5% PuO_2 and a 100 gm lot of UO_2 -20% PuO_2 was completed this month in addition to several kilograms of UO_2 for blending studies. The coprecipitated UO_2 - PuO_2 products were prepared using dilute aqueous ammonia as a precipitant in order to reduce final product surface area as discussed in (1). Results of powder characterization are presented below.

Preparation of several 200 gm lots of PuO_2 utilizing a modified continuous oxalate process is also nearing completion. This is the first of a series of continuous preparation runs to be made under process conditions chosen to give a product of specified surface area ($6 \pm 2 \text{ m}^2/\text{g}$). Earlier runs provided the basic information required for control of product properties. Four additional lots of PuO_2 and UO_2 - PuO_2 will be prepared during the coming month for fabrication into irradiation test elements, for characterization, and for sintering studies.

Surface area measurements were completed on the UO_2 - PuO_2 oxides prepared during this month. The data are summarized in Table I. Sample Pu-3⁽²⁾ is included for comparison.

Table I

Summary of Surface Area Determinations

Sample Identification	% PuO_2	Aqueous Ammonia Concentration (M)	Calcination ^(a) Temperature °C	Surface Area m^2/gm
297-Pu-6	20	14.3	740	7.1
297-Pu-7-L.F.	20	3.5	540	20.7
297-Pu-7-H.F.	20	3.5	740	19.8
297-Pu-8 ^(b)	5	3.5	740	6.4
297-Pu-8 ^(c)	5	3.5	740	6.1
Pu-3	20	14.3	540	17.8

(a) 6% H_2 -94% N_2 atmosphere

(b) Composite of startup and end-of-run product from continuous precipitation

(c) Steady-state product from continuous precipitation run

(1) NUMEC Quarterly Progress Report P-70 (June 1961), page 11.

(2) NUMEC Quarterly Progress Report P-60 (March 1961), page 10.

It is noted that the surface areas of the 297-Pu-7-L.F. and 297-Pu-7-H.F. powders, precipitated with a dilute ammonia solution, are not strongly dependent on the calcination temperature. On the other hand, comparison of 297-Pu-6 with 297-Pu-3, both precipitated with concentrated ammonia, indicates a strong dependence of surface area on temperature. It appears, then, that the combination of ammonia concentration and calcination temperature can be used to obtain and control the desired surface area. Fabricability and sinterability properties of powders prepared under these conditions are yet to be determined.

Surface area measurements were also completed for the 297-Pu-4 and 297-Pu-5 PuO_2 samples. Good comparisons could not be made, however, since repeat measurements showed that these samples are also subject to an apparent surface area rise with storage time⁽²⁾. These powders are being re-examined with the object of finding a suitable pre-treatment to allow attainment of reproducible surface area measurements.

MSA centrifugal particle size distribution measurements employing an ultrasonic field to disperse the powders were completed on the 297-Pu-4 and 297-Pu-5 PuO_2 samples. Comparison of the results with those where overnight shaking was employed indicated that the 297-Pu-5 samples (with the exception of 297-Pu-5-4) were more easily de-agglomerated than 297-Pu-4 powders.

Thermogravimetric studies on the conversion of 297-Pu-4 plutonium oxalate were made employing a heating schedule simulating the conversion processing furnace schedule. These studies are being made to aid in understanding the effects of processing conditions and to aid in optimizing the variables involved.

FABRICATION AND EVALUATION OF FUEL SHAPES

Cold Pressing and Sintering Operations: Samples from two lots of PuO_2 made by continuous precipitation of Pu oxalate were calcined at 350, 420, 490, 560, and 760°C. Results of pelletizing and sintering of the samples calcined from 350 to 560°C were previously reported⁽²⁾. During this period, the samples calcined at 760°C were processed. These two samples were designated 297-Pu-4-5 and 297-Pu-5-5. As had been expected from the relative surface areas, these powders were more stable in air than the powders calcined at lower temperature; vacuum drying at 70°C and 29.5 inches Hg vacuum resulted in weight losses in the order of 4 mg/gm as compared to losses of 6 to 17 mg/gm for the lower temperature calcined powders. Dry pressing of the 297-Pu-4-5 and 297-Pu-5-5 powders resulted in pellets of higher green density than had been achieved with any other previously prepared PuO_2

(2) NUMEC Monthly Progress Letter P-33 (July 1961)

powders (6.7 to 7.24 gm/cm³ as compared to a maximum of 6.23 gm/cm³ for the lower calcined powders). Although the green density was higher, there was no noticeable increase in the sintered densities; the sintered densities on most of the pellets in these two powder lots were 10.5 to 10.8 gm/cm³. All sintering runs were performed at 1600°C in a 6% H₂-94% N₂ atmosphere for one hour.

To determine if the weight decreases on sintering noted on the 297-Pu-4 and 297-Pu-5 powder lots and previously reported⁽²⁾ (as great as 10%) could be partially attributed to vaporization of PuO₂ or residual Pu₂O₃, samples are being sintered for longer time periods at 1600°C. To date, samples have been sintered at 1, 5, and 10½ hours. The weight decrease appears to occur within the first hour of sintering; thus, vaporization cannot be considered a significant factor. Longer sintering runs will be performed to verify these results.

Investigations of binder additives to the PuO₂ to improve the handling and pressing characteristics are being performed. No satisfactory procedure using Carbowax has been found. To date, water has been found to be the best additive to PuO₂ to improve pressing characteristics. In most cases the moisture adsorbed from the atmosphere has been sufficient; however, this is only true for the powders calcined at the lower temperatures (up to 560°C). The powders calcined at 760°C are less adsorbant and additional water must be added. Further work involving the addition of polyvinyl alcohol is underway.

Metallography and X-ray Diffraction: Examination by x-ray diffraction has been continued on a series of PuO₂ samples which showed unexpected metallographic features. Only one face centered cubic phase was detected in the powder prior to sintering. A second face centered cubic phase has been detected in PuO₂ pellets sintered at 1600°C in N₂/H₂, Ar/H₂ and vacuo. A third phase has also been observed in the N₂/H₂ and vacuum sintered pellets but not in Ar/H₂ sintered ones. Lattice parameter measurements of the face centered cubic matrix and face centered cubic precipitate are currently being evaluated. Work on installation of equipment is continuing in the decontamination box, welding box, and centerless grinder box. Experiments utilizing liquid nitrogen as the coolant during centerless grinding are being carried out. The welding box (Blickman type) is being modified for attachment to the decontamination box.

REACTOR PHYSICS AND ENGINEERING PARAMETRIC STUDIES

An analysis has been made of the resonance integral data obtained by the Bettis group⁽⁴⁾ in order to attempt to resolve the discrepancy between

(4) Nuclear Sci. and Eng. 9, 421 (1961)

resonance escape theory and experimental data. The Bettis group found that the effective resonance integral of an 0.387 inch diameter UO_2 rod was 1.30 times greater than that measured for a metallic uranium rod of the same dimensions. (5) This differs appreciably from the 1.41 factor obtained by Hellstrand (2) and used in the development of the NUSURP procedure. The 1.30 factor has now been used in the NUSURP development, and it has been found that experiment and theory are in essential agreement for this one point. Further analysis may show that the NUSURP development can be used to yield consistent values for the effective resonance integral for all fuel rod sizes of interest.

Programming of NUSURP for the RPC-4000 machine is continuing satisfactorily. Completion of machine changeover will be accomplished during the next period.

Very truly yours,

K. H. Puechl / L.f.f.

Karl H. Puechl
Deputy Director
Plutonium Program

lkp

(5) J. Appl. Phys., 28, 493 (1957)

EXTERNAL DISTRIBUTION

U. S. Atomic Energy Commission, New York Operations Office
M. Goldberg, Contracts Division
S. Zirin, Contracts Division
M. Balicki, Research and Development Division
O. Topper, Research and Development Division
H. S. Potter, Patent Group

U. S. Atomic Energy Commission, Chicago Operations Office
B. Anderson

U. S. Atomic Energy Commission, National Laboratories
F. C. Moesel, Office of Technical Information - Washington (2)
J. M. Simmons, Division of Reactor Development - Washington (3)
M. Whitman, Division of Reactor Development - Washington
T. O. Ziebold, Division of Reactor Development - Washington
Reports and Statistics Branch - Washington
F. Foote, Argonne National Laboratory
H. Young, Argonne National Laboratory (4)
A. Squire, Bettis Atomic Power Laboratory
E. A. Evans, Hanford
J. Musser, Hanford
I. D. Thomas, Hanford
O. J. Wick, Hanford
W. K. Woods, Hanford
W. Cashin, KAPL
R. D. Baker, Los Alamos Scientific Laboratory
L. B. Jones, Mound Laboratory
Office of Technical Information Extension, ORNL

U. S. Atomic Energy Commission, Contractors
W. O'Leary, Allis Chalmers Manufacturing Company
R. W. Dayton, Battelle
W. Duckworth, Battelle
R. H. Gale, Combustion Engineering
L. A. Matheson, Dow Chemical Company
G. Spaulding, Electric Autolite Company
A. Strasser, Nuclear Development Corporation of America
Olin Mathieson Chemical Corporation
R. W. Hartwell, Power Reactor Development Company

NUMEC DISTRIBUTION

R. J. Atkins	A. Kasberg
A. Biancheria	H. Krake
F. Cain	A. L. Maharam
C. S. Caldwell	O. Menis
B. M. Cinai	L. P. Pepkowitz
J. Eck	K. H. Puechl
F. Forscher	P. Rey
R. Frumerman	D. G. Ross
H. J. Garber	Z. M. Shapiro
E. E. Garcia	J. Stoner
O. S. Gray	M. M. Turkanis
J. Goodman	B. Vondra
E. K. Halteman	S. A. Weber
R. M. Horgos	A. M. Weis
R. A. Jaroszeski	Library (5)
L. J. Jones	
W. Judd	