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NUMEC P-39

March 5, 1962

Mr. Morris Goldberg, Director
Contracts Division
New York Operations Office
U. S. Atomic Energy Commission
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 FEBRUARY 1962

Gentlemen:

NUMEC herewith transmits the Monthly Progress Letter for February 1962 covering work performed under Contract AT(30-1)-2389.

Major effort during this period has been directed towards the in-pile testing program. Component PuO_2 and UO_2 powder blending trials are being continued as are characterization studies of coprecipitated powders. Also, the alpha boxes for the hot cell are being outfitted, and the Central Research manipulators obtained from excess property for the hot metallographic cell are being repaired and modified. The Preliminary Request for Irradiation Services (AEC Form 21) has been approved for the irradiation of the series of short exposure rabbit capsules. Reactor space has been obtained in the Westinghouse Testing Reactor (WTR), Waltz Mills, Pennsylvania. Preliminary discussion with WTR Project Engineers has resulted in approval of the test specimen and general approval of the capsule design.

DISCLAIMER

The powder blending studies have been continued in order to develop procedures that yield essentially homogeneous UO_2-PuO_2 pellets from mechanically blended component powders. Autoradiographs of pellets give indication that powder blending by wet ball milling produces a nearly homogeneous solid solution UO_2-PuO_2 composition throughout the pellet.

The effect of further heat treatment on powder characteristics following 80 minute reduction at 740°C of UO_2-5 w/o PuO_2 powders prepared by co-precipitation with 14.5 molar ammonia has been investigated. The results indicate that the surface area of these powders has been stabilized; i.e., that further heat treatment (up to 70 minutes) at 740°C does not substantially reduce the surface area, nor does further heat treatment (up to 35 minutes) at 780, 820, or 860°C. While surface area was not affected by this further heat treatment, agglomerate air permeability particle size increased five-fold between 780 and 860°C.

Two additional 350 gram lots of UO_2-5 w/o PuO_2 have been prepared and characterized in order to determine the effect of hammer milling. The progressive changes in the particle size distribution as a result of such milling are now being evaluated.

Particle size distributions of several plutonium (IV) oxalate and plutonium dioxide samples have been measured with the Sharples micromerograph air sedimentation equipment over a wide range of gas feed pressures to optimize conditions for these materials. Feed pressures of 100-125 psig yielded satisfactory dispersion of all powders although some dependence on powder history was observed. Using a feed pressure of 125 psig, comparison of precursor oxalates (aged in an air atmosphere) with their corresponding oxides indicates that particle degradation occurs during conversion. Studies are now in progress using fresh oxalate samples.

Comparative PuO_2 moisture pickup measurements using a recording thermobalance have shown that intermediate surface area (10 M^2/gm) oxide absorbs 90% of its equilibrium moisture content after one hour exposure to ambient air at 76% relative humidity; in comparison, high surface area (60 M^2/gm) oxide requires 6-7 hours exposure before 90% equilibrium moisture content is attained. From the limited results obtained to date, it appears that the equilibrium moisture content is proportional to the surface area at 76% relative humidity (0.3% equilibrium moisture for 10 M^2/gm powder and 1.8% equilibrium moisture for 60 M^2/gm powder).

In preparation for alternate peroxide and oxalate preparation routes for UO_2-PuO_2 mixed oxide, a wide variety of studies on UO_2 preparation have been completed. Specifically, 16 runs have been made using the uranyl peroxide route in order to study the effect of reactant concentrations, feed acidity, and excess reactant level. Two additional runs were made using UO_4 seed nucleation to improve solids filterability and to minimize losses. Four uranyl oxalate preparations were also made using oxalic acid and diethyl oxalate as reactants. Characterization data on these precursor and oxide materials is now being developed. The laboratory scale preparation glove box wherein similar studies will be performed on PuO_2 is in the latter stages of outfitting and checkout trials with non-hazardous materials have been initiated.

Development of alternate methods for agglomerating and shaping spherical PuO_2 particles is continuing. Satisfactory particle shapes have been obtained by a dry ball-mill agglomerating technique. This product has been sintered to a high density at $1600^\circ C$, and evaluation is now underway.

Impurities have been found in the cold entrance zone and the furnace tube zone adjacent to the sintering zone in one of the high temperature sintering furnaces. Analysis of a sample taken from the cold zone showed the presence of cadmium, boron, manganese, gallium, and lead. Presumably, these impurities had been volatalized from pellets. Analysis of a sample taken from near the sintering zone showed the presence of molybdenum, silicon, uranium, nickel, iron, and a trace of plutonium. Subsequent spectrographic analysis of the furnace components (wire, tube, grain, and cement) showed that these materials were free of the offending high cross section elements. It has now been established that these materials were present in significant amounts in the plutonium nitrate as received. This is now being discussed with the supplier. The sintering furnace has now been cleaned of all loose powdery material, and molybdenum sintering boat covers are now being used to minimize furnace contamination.

Economics studies on near-thermal reactor systems indicate that plutonium fuel cycle costs are appreciably below uranium fuel cycle costs even when analyses are based on assuming an ideal burnable poison for the uranium systems. This analysis further substantiates previous conclusions that plutonium value is greater than uranium value in such reactor systems. Since a major fuel cycle cost item for the plutonium systems is the inventory charge, studies are now underway to determine the effect of utilizing some inert material such as ZrO_2 in the fuel elements. The use of such inert material should allow reduction in inventory charge and will not result in reduction in core life (i.e., in MWD/Tonne allowable irradiation) since the irradiation limit is probably determined by materials limitations and not by reactivity limitations. If the results of these economic studies turn out as contemplated, some materials development effort will be directed towards the use of inert materials in fuel elements.

Very truly yours,

Karl H. Puechl

Karl H. Puechl
Acting Director
Advanced Materials Center

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