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COATED FUEL PARTICLES

by

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For presentation at

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COATED FUEL PARTICLES

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INTRODUCTION

The original objective of the work on coated fuel particles at the Nuclear Materials and Propulsion Operation of the General Electric Company (GE-NMPO) was to prepare coated fuel particles less than twenty microns in diameter and to incorporate them in BeO to yield a fuel element in which fuel-matrix reactions, grain growth, and fission product release would be inhibited up to 1650°C.

At the request of the Commission, the objective was modified early in January 1962 to prevent overlap of efforts at GE-NMPO with those of the Washington Research Center of W.R. Grace and Company under USAEC Contract AT(30-1)-2884. W.R. Grace and Company has assumed responsibility for study of the preparation and coating of colloidal fuel particles (0.003 to 0.5 microns). At GE-NMPO studies in the preparation and coating of particles in the colloidal size ranges have been discontinued, and responsibility for evaluating certain W.R. Grace sol materials in a ceramic matrix has been assumed. Therefore, the current objective at GE-NMPO is to prepare coated fuel particles from 0.5 to 50 microns in diameter and to evaluate the usefulness of particles 0.003 to 50 microns in diameter in BeO.

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PARTICLE PREPARATION

Electrodialysis - Before preparation of colloidal materials was discontinued at GE-NMPO, sols of ZrO_2 and UO_2 were prepared by the electrodialysis technique from water solutions of $ZrCl_4$ and UO_2Cl_2 respectively. The ZrO_2 sol particles, shown in Figure 1, were approximately 50 millimicrons in diameter and were probably made up of subunits too small to be resolved at a magnification of 100,000X. The UO_2 sol particles, shown in Figure 2, were also approximately 50 millimicrons in diameter, and some of the particle substructure was resolvable at 150,000X magnification. Results of x-ray diffraction examination of the UO_2 particles indicated an average crystallite size of 25 millimicrons.

Plasma Flame - To achieve flexibility in composition of fuel particles for coating studies above the colloidal size range, the preparation of high density spheres in the plasma flame was undertaken. Plasma flame equipment manufactured by Thermal Dynamics Corporation was used for the spheroidization work. The Thermal Dynamics equipment included three Miller SR 600 rectifiers with 60 KW capacity on continuous operation, a Model B Control Console, and an F-40 torch. The torch was assembled with a particle collector, as shown in Figure 3, to permit spheroidizing of toxic materials. A mixture of argon and helium was used to provide the plasma flame. The particles to be spheroidized were introduced into the plasma flame near the exit end of the gun by a flow of nitrogen gas through a DeVilbiss suction feed cup assembly, which contained the particles.

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Neg. 487C

100,000 X

Fig. 1 - Spherical ZrO_2 particles in a filmy network

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150,000 X

Fig. 2 - UO₂ sol particles

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Fig. 3 - Schematic diagram of fuel particle collecting system
with plasma torch in place

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Spheroidizing of F-116 particles (45 w/o UO_2 -55 w/o Y_2O_3) in the -270 +325 U.S.B.S. sieve number size range is carried out at settings of 600 amperes and 32 volts with gas flows of 30 scfh argon and 10 scfh helium to the plasma gun and 3 scfh of nitrogen through the particle feed system.

A photomicrograph of spheres obtained by spheroidizing F-116 particles in the 37 to 44 micron size range is shown in Figure 4. It may be noted that the spheres are quite dense and that the number and size of voids is fairly small. Whether or not such void spaces are desirable may depend upon the reactor application of the particles and should be a subject of future investigation.

A 7.5 KW Lepel Induction Generator is being modified to generate power at 4 megacycles per second. The generator will be used with an assembly similar to that shown schematically in Figure 5. An induction coil of a few turns surrounds a quartz tube and serves to transfer energy from the generator to an ionized gas passing downward through the tube. The plasma is initiated by heating a graphite or refractory metal in the RF field and continues to propagate against the gas flow. The fuel particles are dropped through the quartz tube and are spheroidized while passing through the plasma zone.

PARTICLE COATING

Wet Chemistry - The objective of the wet chemistry method is to coat spherical fuel particles with a hydrous oxide or with a metal organic

Fig. 4 - Photomicrograph of F-116 spheres

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Fig. 5 - Temperature isotherm configuration
in induction coupled plasma

compound which can be thermally converted to the desired oxide. Such a coating might be attained by adding a precipitating anion to a solution of a salt of the desired oxide or by homogeneous hydrolysis of a salt of the desired oxide. In either case the fuel particles are maintained in suspension by stirring of the solution.

Studies of the first method have involved precipitation of aluminum 8-hydroxyquinolate or zirconium cupferrate in stirred solutions containing spherical fuel particles. To date only very thin coatings have been deposited by this method, and only a fraction of the particles in the mixture have been coated.

Thicker coatings and a higher fraction of coated particles have been obtained by slow hydrolysis of beryllium acetate in solution either by addition of urea or by distilling off acetic acid. Thin coatings have been obtained also from the urea hydrolysis of aluminum sulfate. Coatings obtained from beryllium acetate are shown in Figure 6.

In attempts to encourage precipitation from solution on the particle surface at the expense of formation of precipitate in the bulk of the solution, a surface active agent, Separan NP10, has been used in several coating experiments. Separan NP10 is a polyacrylamide which adheres well to oxide surfaces. Thus it was hoped that the surface active agent would attach itself to the surfaces of the fuel oxide spheres and then attract the precipitating coating material to itself. Results obtained thus far are inconclusive and further studies are being pursued.

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500 X

Fig. 6 - Fuel particles coated by hydrolysis ^{of} ~~by~~ beryllium acetate

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Vapor Reaction Processes - Two types of apparatus for vapor reaction coating are in the process of development. The first type involves a centrifugal bed and the second a counter-flow system. In either type of apparatus the desired coating oxide is produced in the coating chamber by the reaction of a metallic chloride with water vapor as shown in the following chemical equation:

The required water vapor can be produced in the reaction chamber by the reaction,

The centrifugal bed apparatus is shown schematically in Figure 7. The reacting gases are brought to the reaction chamber in two separate paths as shown. Along one path the gases enter tangentially through the wall of the reaction chamber, and the fuel particles are thus caused to spiral upward. The increase in hydraulic diameter with height causes a gradual decrease in velocity of the spiraling gases and thus permits the particles to drop from the gas stream alongside the tube projecting from the floor of the chamber. Therefore the particles pass repeatedly through the coating zone near the bottom of the chamber.

Using this apparatus, F-116 spheres in the -270 +325 mesh size range have been coated with approximately 7 microns of Al_2O_3 in 4 hours at approximately 1000°C. The **coated particles** were tested

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Fig. 7 - Centrifugal bed coating apparatus (AS-104)

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in 8 molar HNO_3 for 8 hours at 70°C . At the end of 8 hours only a few of the fuel cores had been attacked by the HNO_3 . The coated particles before and after testing are shown in Figures 8A and 8B respectively. Major effort in vapor reaction coating is being centered on the centrifugal bed process.

The counter flow apparatus is shown in Figure 9. As shown in the figure, the particles are raised by a gas lift and then dropped through the hot coating zone. As the particles drop to the bottom of the apparatus, they are again lifted for another cycle through the coating zone. Using a power supply capable of producing 600 amperes direct current at 40 volts, temperatures up to 1800°C have been achieved in the particle heating portion of the resistance heated graphite tube. The lift has been used successfully to lift -200 mesh ZrO_2 particles for cycling through the apparatus.

PARTICLE EVALUATION

Thus far evaluation of coated and uncoated fuel particles has been largely confined to microscopic and metallographic examination. However, as mentioned previously, acid leach testing has been started on fuel particles coated with Al_2O_3 in the centrifugal bed apparatus.

PLANS

Particle Preparation - Spheroidization of fuel particles will be carried out in the inductively coupled plasma apparatus and kilogram quantities of particles in several size ranges between 1 and 50 microns will be prepared.

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500 X

Fig. 8A - Photomicrograph of F-116 spheres coated with Al₂O₃

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Fig. 8B - Photomicrograph of Al₂O₃-coated F-116 spheres after acid leach test

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Fig. 9 - Schematic view of counter flow column coating apparatus (AS-68)

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Coating - Studies of particle coating by the wet chemistry method will be continued with emphasis on the effects of surface activation of the particles and on converting and densifying coatings already obtained. Emphasis on coating by vapor deposition methods will be directed toward completion of the centrifugal bed coating apparatus and on studies of the coating of fuel spheres with alumina.

Evaluation - Fuel particles coated successfully will be tested initially by thermal shock, fuel leach at room temperature with a leaching agent which does not attack the coating, and air oxidation at elevated temperatures. Particles which pass the initial tests will be studied for their suitability for incorporation in a BeO matrix.

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