D R A F T (3/28/73) Conf-730601--2

50-word abstract for the 11th Biennial Conference on Carbon

REMOTE COATING OF HTGR FUEL PARTICLES*

W. J. Lackey, W. H. Pechin, and J. D. Sease

Equipment and processes are being developed to allow remote coating of 350-μm-diam $(Th_{0.81},{}^{233}U_{0.19})O_2$ recycle fuel particles in the TURF pilot plant at the rate of 10 kg of heavy metal per day. Carbon coatings and perhaps SiC coatings will be required. Particles having the desired coatings have been produced in 1- to 3-kg batches in a fluidized-bed 5-in.-diam prototype furnace.

*Research sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation.

D R A F T (3/28/73)

Extended Abstract for the 11th Biennial Conference on Carbon

REMOTE COATING OF HTGR FUEL PARTICLES

W. J. Lackey, W. H. Pechin, and J. D. Sease

INTRODUCTION

High-Temperature Gas-Cooled Reactors (HTGR's) have the potential of lowering power costs and improving fuel utilization if fuel recycle capability is established. The HTGR fuel cycle begins with fuel containing thorium and ^{235}U. As a result of neutron capture and subsequent radioactive decay, the thorium is converted to ^{233}U. Basically the fuel recycling process consists of separating the bred ^{233}U from the fission products and refabricating the ^{233}U into a suitable form for reinsertion into the reactor. The proposed recycle fuel consists of microspheres of $(Th_{0.81},{}^{233}U_{0.19})O_2$ about 350-μm-diam that are coated with carbon and, after mixing with carbon-coated ThO_2 microspheres, formed into fuel sticks about 5/8-in.-diam and 2- to 4-in. long. These sticks, after carbonizing and annealing, are loaded into holes in hexagonal graphite blocks which are stacked to form the reactor core. When fuel cycle equilibrium is reached, about half the fissile material needed for reactor refueling is provided by the ^{233}U.

The objective of the National HTGR Fuel Recycle Development Program[1] is to develop recycle technology so that commercial plants for refabrication of HTGR fuels can be built and operated economically. To accomplish this objective, it is necessary to perform equipment and process development involving engineering, design, and testing. The recycle fuel must be handled in heavily shielded facilities because of the presence of some ^{232}U which has ~~daughter~~ decay products that emit high-energy gamma radiation. The particular portion of this program described here is concerned with coating of the recycle fuel particles. Our objective is to develop remote coating technology and to demonstrate this technology by operation in 1979 of a pilot plant located within the Thorium-Uranium Recycle Facility (TURF) hot cells.

The particle coating task includes particle inspection and particle handling in addition to the coating operation. The particles must be sampled and inspected. They must be classified to the proper size range and shape separated, and provisions must be made for particle transfer, storage, and blending. Only in the case of particle inspection can the operations be performed in glove boxes; the remaining operations involve kilogram rather than gram quantities of fuel, and thus the shielding provided by a hot cell is required.

---

[1] National HTGR Fuel Recycle Development Program Plan, Oak Ridge National Laboratory and Gulf General Atomic, ORNL-4702 (August 1971).

PARTICLE COATING

Fluidized bed-type particle coating equipment and processes are being developed for remote coating of recycled HTGR fuel. Porous carbon coatings, which provide void space for accommodation of fission products and which protect the outer coating from recoiling fission fragments, are applied by decomposition of acetylene diluted with helium. Dense, strong, isotropic carbon coatings and perhaps SiC coatings will be required for fission product containment. The high-density pyrolytic carbon coating is applied by decomposition of propylene. Coatings of SiC are formed by decomposition of methyltrichlorosilane, CH_3Cl_3Si, diluted with hydrogen. Temperatures range from 1000 to 1700°C, depending on gas type and coating properties desired.

The reference recycle fissile particle is of the BISO-type and consists of a 350-μm-diam sol-gel $(Th_{0.81},{}^{233}U_{0.19})O_2$ kernel coated with buffer and isotropic carbon coatings each about 100 μm thick. For particles of this type, about 10 kg of heavy metal will be coated daily in the TURF pilot plant. Such particles have been produced numerous times in 1- to 3-kg batches in the 5-in.-diam prototype remotely operated coating furnace which has been described previously.[2–4] Although extensive additional equipment modification

---

[2]R. B. Pratt and S. E. Bolt, Status and Progress Report for Thorium Fuel Cycle Development for Period Ending Dec. 31, 1966, ORNL-4275, pp. 61–78.

[3]F. J. Furman, Jr., J. D. Sease, and A. L. Lotts, "Economics and Technology of High-Temperature Gas-Cooled Reactor Fuel Refabrication," Symposium on Sol-Gel Processes and Reactor Fuel Cycles, Gatlinburg, Tennessee, May 4–7, 1970, pp. 281–308, CONF-700502.

[4]W. J. Lackey, et al., Gas-Cooled Reactor and Thorium Utilization Programs Ann. Progr. Rept. for Period Ending Sept. 30, 1971, ORNL-4760, pp. 45–50.

will be required for fully remote operation, the coater is currently highly automated, versatile, and reliable. Important features of of the coating furnace system are summarized below.

1. Modular design is used to facilitate in-cell maintenance.
2. The total heat capacity of the furnace is low to permit accurate temperature control during rapid change in conditions in the coating region and to allow rapid cooldown to facilitate particle unloading and routine maintenance.
3. Exhaust pump allows variation of the furnace pressure if desired, and permits absolute filtration of exhaust gas.
4. The flow of combustible gases is controlled without routing these gases to the remote control panel.
5. The hydrocarbon and diluent flow rate can be programmed to maintain a constant gas flux throughout the coating run.
6. Numerous temperature, pressure, flow, and combustible gas monitors are interlocked within the system to protect both the operator and equipment.
7. Hydrogen chloride produced during deposition of SiC coatings is removed from the effluent gas via a caustic scrubber.

Although numerous types of gas injectors and coating chambers have been used for bringing the coating gases into contact with the bed of particles in the coating furnace, most of our experience has been with a single-inlet, 30° included-angle conical gas distributor. With reference to the buffer coating process, statistically designed experiments have been conducted where the variables of interest were acetylene flow rate, type of diluent gas, diluent gas flow rate, temperature, time, kernel size, standard deviation of kernel size, and charge weight. Responses of interest were coating thickness, particle-to-particle variation in coating thickness, coating density, efficiency of utilization of input carbon, and various system responses such as the extent of soot formation and the influence of soot on the behavior of the soot filter. In addition to recently achieving a vastly improved understanding of the buffer coating process that will allow us to confidently deposit buffer coatings having the required properties in the future, we were successful in finding a new set of operating parameters that reduced the within-batch coating thickness standard deviation from about 25% of the mean coating thickness to about 14%. For the reference buffer thickness of 100 μm, this improvement in coating thickness uniformity means that instead of 1% of the particles in a batch having coatings thinner than 48 μm, only 1% of the coatings will be thinner than 66 μm. Since particles with thin buffer coatings are more likely to fail during irradiation, this process improvement should yield fuel significantly superior in irradiation performance. Not only did the new set of operating conditions yield coatings more uniform in thickness from particle-to-particle, but the coatings were of the desired density of 1.1 ± 0.2 g/cm³ and the process was rather insensitive to the quantity of material being coated. Charges as large as 3200 g of ThO_2 kernels were successfully coated. For charge weights in the range 800 to 3200 g the coating efficiency was 48% independent of charge weight, and the density was rather insensitive to charge weight for the conditions used.

The improved buffer coating process was accomplished by increasing the diluent flow rate, the acetylene flow rate, and temperature. The single most important factor was the increased diluent flow rate. Higher diluent flow rates decreased the particle-to-particle variation in coating thickness. However, unless the acetylene flow rate and/or temperature are also increased, coating densities are unacceptably high. Higher gas flow rates apparently lead to better mixing of the spouting particle bed and, therefore, more uniform coating thicknesses. The use of helium as a diluent is preferable to argon since coating densities are lower when helium is used, which means that higher diluent flows can be used without producing coatings that are too dense.

These experiments not only gave us a better qualitative understanding of the buffer process, but considerable success in quantitatively understanding the process was achieved. Phenomenological equations were developed for predicting the mass of carbon deposited, the coated particle volume and density, and the standard deviation of coating thickness. The independent variables in these expressions were charge weight, time, acetylene flow rate, mass ratio of diluent flow to acetylene flow, standard deviation of kernel diameter, and kernel diameter.

The effect of varying the included angle of the single inlet conical-type gas distributor was investigated. For angles of 30, 90, and 135° the performance, as measured by uniformity of coating thickness and coating efficiency, improved considerably as the cone angle was decreased.

We discovered that for a given buffer coating run the coating density was greater for the thin coatings. The fact that the thinner coatings are more dense means that the average buffer coating thickness will have to be about 5% larger than would be required if there were no relationship between thickness and density.

A limited effort was directed toward investigating the effect of temperature and deposition rate on the properties and structure of SiC coatings applied using the prototype coater. It was shown that as the deposition rate is increased, there is a decrease in coating density and an increase in surface roughness. Both of these effects are expected to result in weaker coatings.[5] Coatings were

[5]A. G. Evans, C. Padgett, and R. W. Davidge, "Strength of Pyrolytic SiC Coatings of Fuel Particles for High-Temperature Gas-Cooled Reactors," J. Am. Ceram. Soc. 56(1), 36–41 (1973).

deposited at a rate of 0.18 μm/min. which were dense and fine grained and appeared to be of excellent quality.

PARTICLE INSPECTION

Fuel kernels and coated particles must be routinely and rapidly inspected for size, kernel density, coating density, coating thickness, and coating anisotropy. Other properties that may require routine or periodic measurement are crushing strength, impurity content, sphericity, and fraction of exposed fuel as a result of defective coatings. The most important single measurements in particle inspection is the diameters of the bare kernel and the coated particle after application of the various coatings. Our current concept for the TURF demonstration involves two different methods of performing the measurement—use of an automatic particle size analyzer and measurement of particles from microradiographs.

A second-generation particle size analyzer has been designed and fabricated and is currently in the final stages of testing and calibration. It is an electronic instrument and makes use of the light blockage principle for counting and measuring the diameter of particles at rates exceeding 2×10^4 particles per minute. The analyzer will be used primarily as a process control means, in that a sample from every coating run will be counted and measured. The instrument has 1000 channels of memory and is capable of direct interfacing with our PDP3/E computer for the statistical analysis and printout of the data. Preliminary results indicate that the mean particle size for a batch reproduces to less than 1 μm.

The measurement of particle diameter from contact microradiographs will be used only as a quality assurance measure in that radiographs of blends from several coating batches will be made and measured to ensure that such blends meet specifications for the mean and standard deviations of particle diameter, coating thickness, etc. Measurement of microradiographs has been improved by adding a digital output to a split-image microscope eyepiece. A PDP8/E digital computer has been mated directly to the eyepiece output to eliminate all intermediate data handling. With this arrangement, it is possible to determine the kernel diameter and coating thickness(es) of 50 particles and to have the required statistics calculated in a total time of 10 minutes or less.

As an aid to statistical studies and to assist in the calculation of daily experimental results, we have begun a program of logging all coating data into a time-sharing computer system. Input data consists of material properties, coating process parameters, analytical results such as particle density and wt % carbon, and radiographically determined values for the coating thickness and volume of coating per volume of kernel. Using these data the computer calculates coating density by three methods, relative coating thickness standard deviation, coating efficiency, and numerous other responses of interest for each coating run.

PARTICLE HANDLING

Particle handling involves screening, shape separation, weighing, batching, sampling, storage, and transfer. Most commercial materials handling equipment is not suitable, since either it is designed to handle ton quantities or it is laboratory equipment and thus not readily adaptable to remote or automatic operation. All particle handling devices and storage hoppers will be connected by pneumatic or gravity transfer lines. The handling techniques planned for TURF are currently either being used in normal laboratory operations or are being developed. A test stand for use in developing particle handling equipment has been operational for several years. The stand includes gravity and pneumatic transfer lines, transfer line valves and connecting storage hoppers, material inventory monitors, and feed devices. The same basic hopper design is used throughout the system, but several different valve designs are being used because of the different functions to be performed. For material inventory monitors the test stand contains a capacitor liquid level probe and a strain-gage bridge circuit.

Classification of microspheres includes the separation of both nonspherical particles and over- and undersized particles. A shape separator to remove nonspherical particles is a flat-plate vibratory feeder with the plate tilted slightly downward in respect to the

~~direction of feeding action and~~ tilted also 90° to this direction. For size classification we have used a modified 18-in.-diam gyratory screen equipped with a self-cleaning attachment. A more detailed description of particle handling equipment is available.[6]

[6]F. J. Furman, J. T. Meador, and J. D. Sease, Microsphere Handling Techniques, ORNL-TM-2782 (March 1970).