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Fabrication and Irradiation Behavior of
Advanced Fuel Sticks for the HTGR*

J. L. Scott, R. B. Pollock, R. L. Hamner, J. M. Robbins,
J. H. Coobs, and W. P. Eatherly

The reference fuel element for the High-Temperature Gas-Cooled Reactor (HTGR) consists of hexagonal blocks of graphite about 14 in. (35.5 cm) across the flats and 31 in. (78.7 cm) long with parallel fuel and coolant holes.¹ The initial fuel consists of fissile particles containing uranium highly enriched in ^{235}U and fertile particles containing only thorium. Both types are prepared as carbide microspheres with coatings of pyrolytic carbon plus SiC to contain fission products. The carbon coatings accommodate fuel swelling and prevent Xe, Kr, and I_2 release and the SiC is a diffusion barrier to Cs, Ba, and Sr. The design of the particles and coatings permits separation of the spent uranium from thorium during head-end processing. The bred ^{233}U in the thorium may then be recovered and recycled into new fuel elements in order to maximize the utilization of fuel resources.²

In fabrication of the fuel elements, the coated fissile and fertile particles are mixed intimately and then bonded together by intrusion of a carbonaceous mix of pitch plus graphite and carbonizing to at least

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100°C in Ar. The fuel sticks, nominally 0.49 in. (12.45 mm) in diameter and 1.0 in. in length, are loaded into the fuel holes of the graphite block. The reference HTGR fuel stick has been tested in numerous irradiation tests by Gulf General Atomic (GGA), OECD Dragon, and Oak Ridge National Laboratory (ORNL), and is considered to be fully developed.

At ORNL work is progressing on advanced HTGR fuel concepts which are more amenable to fuel recycle. These include: (a) sol-gel oxide kernels for both the fissile and fertile particles, (b) fertile particles coated with pyrolytic carbon coatings only, (c) fissile particles made from ion-exchange resins, and (d) fuel sticks with a continuous carbon matrix made by warm molding or extrusion processes. The fabrication of microspheres of UO_2 , $(\text{Th,U})\text{O}_2$, or ThO_2 by various sol-gel processes is developed up to pilot-plant scale.³ In current work the irradiation performance of optimum designs is being emphasized. Likewise, ThO_2 particles with two-layer carbon coatings have been successfully irradiated to burnups higher than that required for the HTGR, but the optimum coating design is yet to be determined. Irradiation tests to define optimum coatings are now underway. One question which must be answered is the rate of release of solid fission products from ThO_2 kernels at burnups to about 8% FIMA.

The study of fissile particles made from ion-exchange resins is relatively new. Fabrication involves loading of uranium as uranyl ions onto either strong-or weak-acid resins, drying, and carbonizing in an inert atmosphere in a fluidized bed. The product consists of spherical particles with up to 4 g U/cm³ of kernel and 30 to 40% porosity. The process is simple and amenable to remote processing. In addition, the existence of the built-in porosity eliminates the need for thick buffer coatings. By appropriate design the same net fuel loadings are achievable with resin-derived particles as those obtainable with dense kernels and thick buffer coatings. Irradiation tests on resin-derived particles have shown excellent irradiation behavior of 60% FIMA provided a thin buffer of porous carbon is applied to decouple the kernel from the coating.

In the current HTGR fuel element the fuel sticks are made from density packed beds of particles occupying ~62% of the volume. By optimizing coating designs, i.e., by use of larger kernels and relatively thinner coatings, fuel sticks can be made which require only about 30 to 40% of the particles.

Two processes that have been used are warm molding and extrusion.

Warm molding involves mixing coated particles, filler graphite, and

binder with a solvent to make a slurry, then drying and molding

The extrusion process is similar except that the forming operation is extrusion instead of molding. Using these techniques we have produced fuel sticks with matrix densities in the range 1.4 to 1.75 g/cm³. Such matrices would distinctly improve the thermal and physical properties as compared to fuel sticks with matrix densities of 0.6 to 0.9 g/cm³ prepared by conventional intrusion bonding. Fabrication studies showed that excellent fuel distribution and dimensional control are achievable. Tests are currently underway to assess the irradiation stability. By proper control and with the use of needle-coke graphite fillers, the dimensional stability throughout life should match closely that of the graphite block containing the fuel sticks.

In addition to the fabrication development work described above, a number of irradiation tests of conventional HTGR fuel sticks have been carried out in the HFIR reactor, where maximum neutron fluences are achieved 12 to 25 times faster than in the HTGR itself. Results showed that bonding matrices made with a variety of carbonaceous filler materials of 8×10^{21} neutrons/cm² ($E > 0.1$ MeV) and coal-tar pitch survive intact to the full HTGR fluence and the overall

dimensional stability of the fuel sticks is governed by the design of the particle coating and the density of the outer pyrolytic carbon. Coatings with intermediate SiC layers shrink less than those without. For the latter the dimensional changes are inversely related to the density of the outer coating. Good dimensional control ($< 3\%$ shrinkage) is achieved either by using SiC barrier layers or by use of coatings with densities greater than 1.90 g/cm^3 .

In conclusion, irradiation testing of conventional HTGR fuel sticks has verified their excellent irradiation behavior and clarified desirable properties of the coated particles used in making the sticks. A variety of coated particles and fuel sticks which are more amenable to ^{233}U recycle than those made by present-day technology have been successfully fabricated. Two particularly attractive fuels are sol-gel oxides or resin-derived kernels. With optimum coating design particle loadings less than 62% can be used. New processes for fabricating such sticks containing $30 \text{ vol } \%$ particles have been developed.

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