MASTER

BNL-NUREG-25715

For presentation at the 14th Carbon Conference, Pennsylvania State University, Uuniversity Park, PA, June 25-29, 1979

CONF-790625--3

FAILURE MECHANISMS IN HIGH TEMPERATURE GAS COOLED REACTOR FUEL PARTICLES*

P. Soo, G. Uneberg, R. L. Sabatini and D. G. Schweitzer
Brookhaven National Laboratory
Upton, New York, 11973

ABSTRACT

BISO coated UO_2 and ThO_2 particles were heated to high temperatures to determine failure mechanisms during hypothetical loss of coolant scenarios. Rapid failure begins when the oxides are reduced to liquid carbides. Several failure mechanisms are applicable, ranging from hole and crack formation in the coatings to catastrophic partic e disintegration.

*This work was performed under the auspices of the United States Nuclear Regulatory Commission.

For presentation at the 14th Carbon Conference, Pennsylvania State University, University Park, PA, June 25-29, 1979.

FAILURE MECHANISMS IN HIGH TEMPERATURE GAS COOLED REACTOR FUEL PARTICLES

P. Soo, G. Uneberg, R. L. Sabatini, D. G. Schweitzer
Brookhaven National Laboratory, Upton, NY 11973

Introduction

This work is concerned with the failure of High Temperature Gas Cooled Reactor (HTGR) fuel particles during thermal excursions up to the point where oxide fuel kernels are reduced to the liquid carbide. The data are of importance in determining the possible mechanisms of failure during hypothetical accident scenarios in which an HTGR is subjected to a loss of coolant event.

Experimental Procedures and Results

Batches of experimental BISO coated UO_2 and ThO_2 fuel particles were obtained from Oak Ridge National Laboratory for this investigation. The UO_2 and ThO_2 kernels are coated with an inner porous carbon layer and an outer dense shell of pyrolytic carbon.

During a typical experiment, a batch of 50 particles is placed inside a small capped graphite crucible and heated in an induction furnace at an initial rate of 2000°C/h to a predetermined temperature. The particles are maintained at this value for a given time and then allowed to cool naturally. Temperatures were measured using an optical pyrometer. Particles were examined after heating with the aid of a scanning electron microscope/microprobe (SEMM).

Figure 1 shows a BISO(UO_2) particle from a batch held for 2 hours at 2400°C. All particles were intact after heating but this particular particle displayed a surface eruption from which uranium was released. A second batch held at a slightly higher temperature (2450°C/0.5 h) also remained intact but one particle was found to have multiple cracks in the outer pyrolytic carbon coating (Figure 2). Uranium was again found to be present in the failure zones.

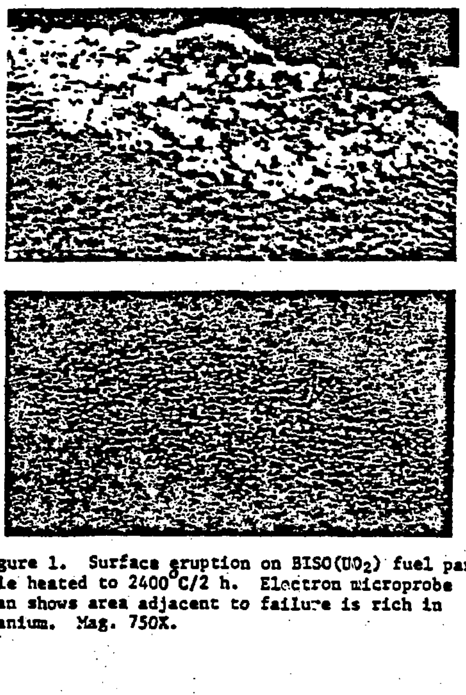

Figure 1. Surface eruption on BISO(UO_2) fuel particle heated to 2400°C/2 h. Electron microprobe scan shows area adjacent to failure is rich in uranium. Mag. 750X.

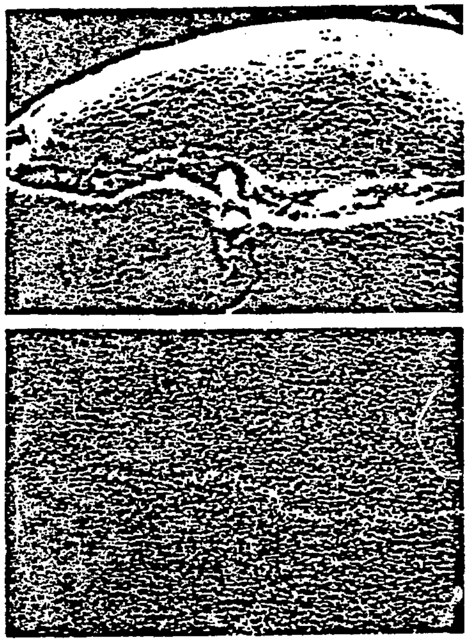

Figure 2. BISO(UO_2) fuel particle heated to 2450°C/0.5 h. Multiple cracks in outer pyrolytic carbon coating; electron microprobe scan showing uranium rich areas. Mag. 325X.

Figures 3 and 4 show that at a test temperature of 2550°C more extensive failure of BISO(UO_2) fuel occurs and approximately 10 percent of the particles displayed some evidence of coating failure. In Figure 3, small nodular masses have broken away from the surfaces of the coating and uranium was found to have escaped from these regions. The failure shown in Figure 4 is in the form of a tulip shaped eruption upon which small white particles are deposited. An SEMM scan shows that the erupted material contains some uranium but the most highly concentrated uranium is present in the white particles.

In this work no BISO(UO_2) particles failed at temperatures below the melting point of UC_2 (i.e. 2400°C). Failure became more prevalent and extensive as the temperature was steadily raised above this value. Based on this observation it is likely that HTGR fuel particle failure is closely associated with the formation of the liquid carbide according to the reaction:

$$UO_2 + 4C \rightarrow UC_2(liq) + 2CO \qquad (T \geq 2400°C)$$

In this temperature range the high CO pressure is believed to rupture the coatings and fuel is expelled through the failed regions. The tulip shaped material in Figure 4 is probably UC_2 and the white particles could be metallic uranium.

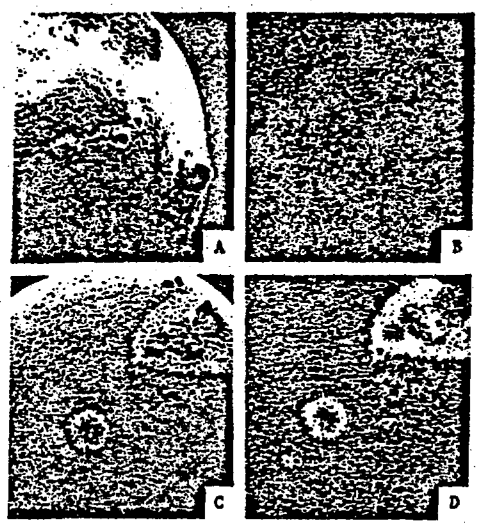

Figure 3. BISO(UO_2) fuel particles heated to 2550°C/1 h. (A) Shows an eruption and fissure and (B) gives an electron microprobe scan of uranium rich areas, mag. 210X. (C) Shows eruptions in a second particle and (D) shows areas rich in uranium, mag. 140X.

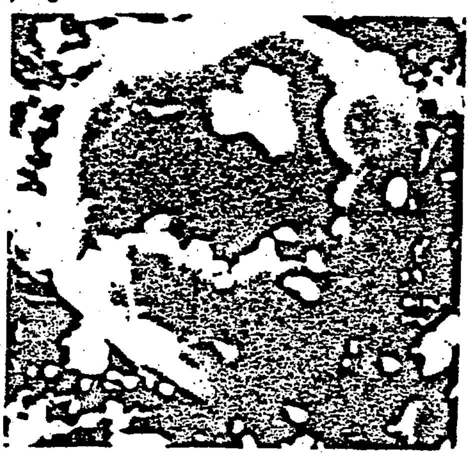

Figure 4. BISO(UO_2) fuel particle heated to 2550°C/1 h. Tulip shaped eruption moderately rich in uranium and white particles are extremely rich in uranium. Mag. 2400X.

A batch of BISO(ThO_2) particles was heated to 2700°C, a temperature slightly higher than the melting point of ThC_2. As the melting point was reached a loud retort was heard from within the induction furnace and a shower of sparks was expelled. On cooling the samples it was noticed that the cap had been blown off the crucible by CO pressure and about 85 percent of the particles were found to have completely fragmented. Of the remaining intact particles several showed a "volcanic" type failure in which the molten fuel was ejected under pressure leaving a round "crater" at the point of initial failure (Figure 5). Figures 1 and 3 show events which could possibly have developed into volvanic failures if the temperature had been high enough to form large quantities of liquid UC_2 and sufficient CO to expel the fuel through small ruptures in the carbon coatings.

Figure 5. BISO(ThO_2) fuel particle heated to 2700°C. Reduction of ThO_2 to liquid ThC_2 causes rapid CO evolution which expels the molten fuel through holes in the carbon coating. Mag. 100X.

Conclusions

BISO coated UO_2 and ThO_2 fuel particles are susceptible to rapid failure if the temperature is raised above the point where the oxide is reduced to the liquid carbide by interaction with the pyrolytic carbon coatings. For temperatures where the amount of carbide is small, failure mechanisms include the formation of small holes or cracks. In other cases liquid carbide appears able to migrate directly through unbroken coatings. At temperatures significantly higher than the carbide melting point, the internal CO pressure inside the particle is sufficiently high to give a "volcano" type failure which leaves an empty carbon shell and a round crater through which the fuel was expelled.

Acknowledgements

This work was sponsored by the United States Nuclear Regulatory Commission.