CONF-881011--40

Received

FEB 2 4 1989

FUEL-SODIUM REACTION PRODUCT FORMATION IN BREACHED MIXED-OXIDE FUEL*

by

CONF-881011--40

DE89 007383

J. H. Bottcher, J. D. B. Lambert, R. V. Strain
Argonne National Laboratory
9700 South Cass Avenue
Argonne, Illinois 60439

and

S. Ukai, S. Shibahara
Power Reactor and Nuclear Fuel
Development Corporation
4002 Narita-cho, Oarai-machi
Higashi-Ibaraki-gun, Ibaraki
Japan

Submitted for Presentation
at the
1988 ANS/ENS International Conference
October 30 - November 3, 1988
Washington, D.C.

*Work supported by the U.S. Department of Energy, Reactor Systems, Development, and Technology, under Contract W-31-109-38.

Fuel-Sodium Reaction Product Formation in Breached Mixed-Oxide Fuel

J.H. Bottcher, J.D.B. Lambert, R.V. Strain (ANL);
S. Ukai, S. Shibahara (PNC, O-Arai, Japan)

The run-beyond-cladding-breach (RBCB) operation of mixed-oxide LMR fuel pins has been studied for six years in the Experimental Breeder Reactor-II (EBR-II) as part of a joint program between the U.S. Department of Energy and the Power Reactor and Nuclear Fuel Development Corporation of Japan. The formation of fuel-sodium reaction product (FSRP), Na_3MO_4, where $M = U_{1-y}Pu_y$, in the outer fuel regions is the major phenomenon governing RBCB behavior. It increases fuel volume, decreases fuel stoichiometry, modifies fission-product distributions, and alters thermal performance of a pin. This paper describes the morphology of Na_3MO_4 observed in 5.84-mm diameter pins covering a variety of conditions and RBCB times up to 150 EFPD's.(1-3)

When a fuel pin breaches and sodium enters it reacts with both the fuel and any existing cesium-fuel reaction compounds. Reaction with fuel is limited by the partial pressure of oxygen and sodium.(4,5) If oxygen in the bulk sodium is <5 ppm, the source of oxygen is the fuel itself. Reaction is generally limited to temperatures below the local boiling point of sodium which is normally 900-1000°C in EBR-II. Sodium will displace cesium in any fuel or fission-product phases, without undue effect on pin performance except to enhance delayed-neutron signals. The extent of FSRP formation is time dependent and ultimately limited by the oxygen source or the dissociation temperature. The resulting circumferential layer of sodium-urano-plutonate is a half-density (5.6 g/cc) fuel that influences pin performance by its different properties.

Conditions change rapidly during the first few days of RBCB operation. Reaction initially occurs under sodium-rich conditions when the fuel-cladding gap conductance may actually be improved.(6) But within 125-150 hours the reaction can consume 80% of the fuel in the first 0.2 mm of fuel up to the dissociation isotherm, producing an adherent dense FSRP around islands of unreacted fuel (Fig.1a). Some porosity due to fission gas is apparent. The FSRP appears amorphous with an absence of grain structure and with spherical voids and occasional lenticular cracks, originating and ending in the product matrix. The 6% volume increase can be locally accommodated without significant increase in pin diameter.

After 1500-3600 hours of RBCB operation the FSRP will essentially be in chemical equilibrium with fuel and sodium. Figure 1b shows the very uniform, 0.17-mm thick FSRP layer in a 9 at.% burnup plenum-defective pin after 152 EFPD's RBCB operation.(2) The layer contains fully reacted FSRP, about 21% fission-gas porosity, and small white fission-product precipitates that are unaffected by the reaction. The fuel volume increase of 6.5% was accommodated with a maximum cladding strain of 0.8%. FSRP morphology in a fuel-column breached pin after 110 EFPD's RBCB operation was very similar to that in the plenum-defective when examined away from the breach (Fig.1c). At the breach where the FSRP layer was exposed to sodium, the fission-gas bubbles were larger, indicative of a stress-free condition and a certain material plasticity (Fig. 1d). But the FSRP layer remained intact and stable and <5 mg of fuel loss was measured during irradiation under the breached-fuel test facility.(7)

The fuel-sodium reaction product layer formed at the surface of breached mixed-oxide fuel pins can be adherent, stable, and protective during long periods of RBCB operation. The product is basically an amorphous, single-phase material that generates fission gas like any other fuel. It is inert to sodium and can act to glue up an open breach. Its influence on fuel-pin performance, particularly thermal behavior (8), is however complex, warranting the continuing study of RBCB behavior in advanced pin designs.

References

1. Lambert, J. D. B. et al., "**Performance of Breached LMFBR Fuel Pins During Continued Operation,**" Proc. BNES Meeting, Nuclear Fuel Performance, March 1985.

2. Bottcher, J. H. et al., "**Breached Pin Performance in EBR-II RBCB Program,**" International Conference on Reliable Fuels for Liquid Metal Reactors, Tucson, Arizona, Sept. 1986.

3. Bottcher, J. H. et. al., "**Breached Oxide Blanket Fuel Behavior in an LMR,**" Trans. ANS, **55**, p. 290, (1987).

4. Adamson, M. G. et al., "**Thermalchemical Aspects of Fuel-Cladding and Fuel-Coolant Interactions in LMFBR Oxide Fuel Pins,**" International Symposium on Thermodynamics of Nuclear Materials, Julich, F.R.G., Feb. 1979.

5. Mignanelli, M. A. and Potter, P. E., "**The Reaction of Sodium with Urania-Plutonia: Behavior of Failed Fuel Pins in a Fast Reactor,**" Proc. of an International Conference, Science and Technology of Fast Reactor Safety, Vol. I, Guernsey, U.K., May 1986.

6. Ukai, S. et al., "**Fuel Performance of Plenum Slit Pin Irradiated in JOYO,**" International Conference on Reliable Fuels for Liquid Metal Reactors, Tucson, Arizona, Sept. 1986.

7. Olp, R. H., et al. "**The Experimental Breeder Reactor (EBR-II) Breached Fuel Test Facility,** Proc. of the Fast, Thermal and Fusion Reactor Experiments, ANS/MS&T, Salt Lake City, Utah, April 12-15, 1982.

8. Lee, M. J. et al., "**Thermal Conductivity of $Na_3(U_{1-y}Pu_y)O_4$, A Preliminary In-pile Determination,**" Trans. ANS, **55**, p. 294 (1987).

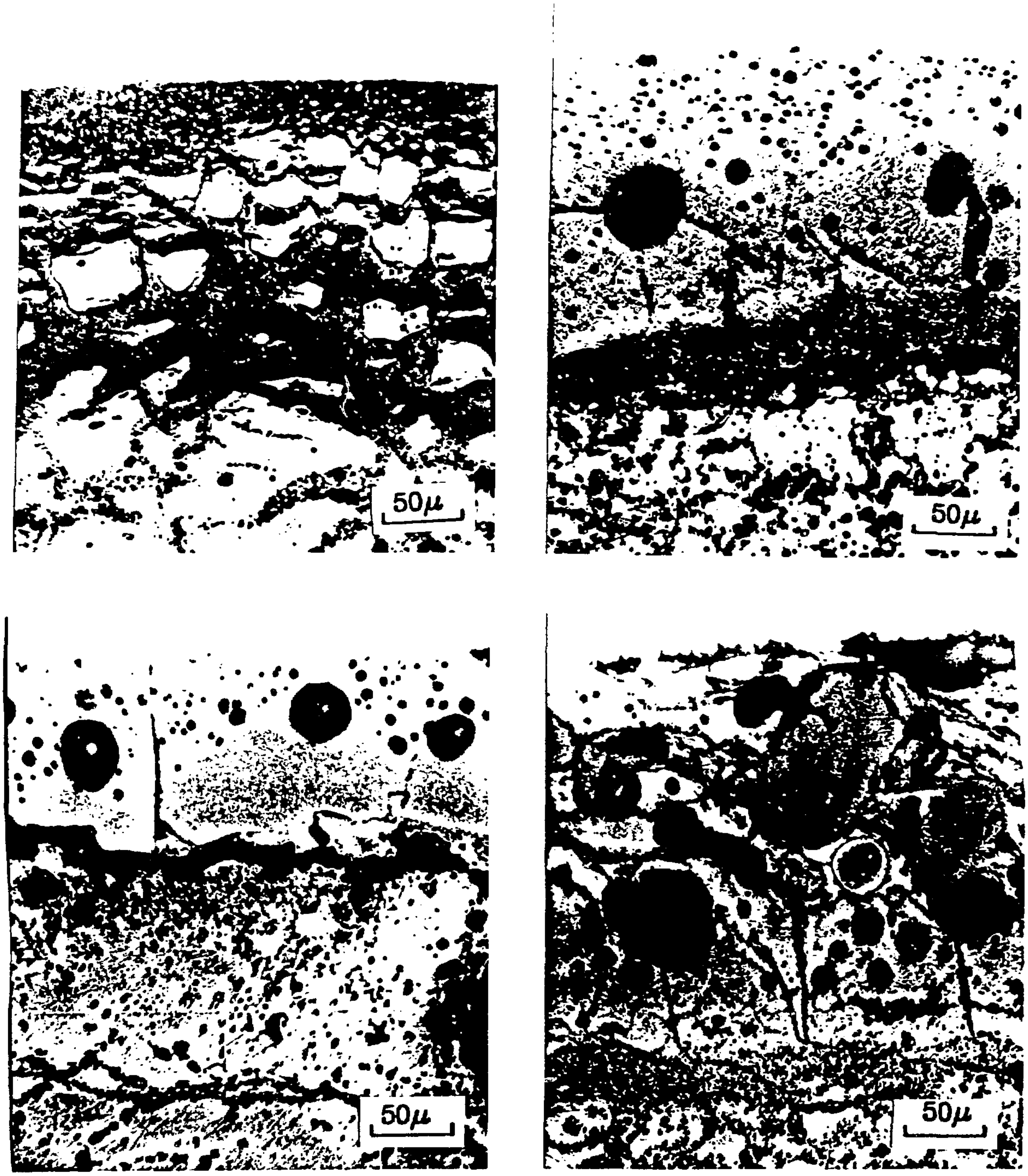

Fig. 1. Na_3MO_4 Morphology with RBCB Operating Time

a. 5 Days

b. 152 Days

c. 110 Days (below breach)

d. 110 Days (at breach)