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**DAMAGE AND FAILURE OF UNIRRADIATED AND IRRADIATED  
FUEL RODS TESTED UNDER FILM BOILING CONDITIONS**

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**ABSTRACT**

Power-cooling-mismatch experiments are being conducted as part of the Thermal Fuels Behavior Program in the Power Burst Facility at the Idaho National Engineering Laboratory to evaluate the behavior of unirradiated and previously irradiated light water reactor fuel rods tested under stable film boiling conditions. The observed damage that occurs to the fuel rod cladding and the fuel as a result of film boiling operation is reported. Analyses performed as a part of the study on the effects of operating failed fuel rods in film boiling, and rod failure mechanisms due to cladding embrittlement and cladding melting upon being contacted by molten fuel are summarized.

**INTRODUCTION**

The primary safety consideration in nuclear reactor design is to ensure that no conceivable accident, whether initiated by a failure of the reactor system or by incorrect operation, will lead to a dangerous release of radiation to the environment. A number of hypothesized accidents must be considered in the design and licensing of light water reactors (LWRs). The extreme examples have been traditionally known as a loss-of-coolant accident (LOCA), in which a part or all the coolant is lost from the active core, and a reactivity initiated accident (RIA), in which a rapid, inadvertent insertion of reactivity results from the ejection of a control rod from the core. Between these two extremes, various off-normal power or cooling conditions may occur, generally termed power-cooling-mismatch (PCM) accidents because of failure of the coolant to successfully remove the heat generated within the core.

Numerous credible single and coincident initiating events that may lead to PCM accidents can be postulated. If departure from nucleate boiling (DNB) occurs during a PCM event, fuel rod damage and subsequent release of radioactivity into the primary coolant system may occur. Current licensing criteria dictate that if a rod is postulated to have exceeded DNB ratio limits, the rod is assumed to have failed. However, as will be shown in this paper, fuel rods can operate in film boiling and incur significant damage before failure actually occurs.

Power-cooling-mismatch experiments are being conducted by EG&G Idaho, Inc., for the Nuclear Regulatory Commission, as part of the Thermal Fuels Behavior Program in the Power Burst Facility (PBF) at the Idaho National Engineering Laboratory to evaluate the behavior of unirradiated and irradiated

LWR fuel rods tested under film boiling conditions. To date, 22 unirradiated, 9 previously irradiated, and 7 fuel rods built with irradiated cladding and fresh fuel have been tested. The results of these tests have been previously reported [1, 2, 3].

This paper summarizes the results from these in-pile tests, with emphasis on the damage incurred by the cladding and fuel during film boiling. Analyses performed on the effects of operation of failed fuel rods in film boiling and on rod failure mechanisms due to cladding embrittlement and cladding melting upon being contacted by molten fuel are summarized.

## EXPERIMENT DESIGN

The tests were performed using pressurized water reactor (PWR) type fuel rods, about 0.97 m long. The rods were tested singly or four at a time in the PBF in-pile test loop, with coolant temperatures and pressures typical of a PWR environment. Each fuel rod was contained in a coolant flow shroud and typically instrumented as illustrated in Figure 1.

Film boiling conditions were established by either reducing coolant flow through the flow shroud while maintaining constant rod power, or increasing rod power while maintaining constant coolant flow. The rods were operated under stable film boiling conditions from 30 seconds up to about 12 minutes with, depending on the test, fuel rod peak powers ranging from about 50 to 78 kW/m. Film boiling operation was terminated by either increasing flow or rapidly reducing fuel rod power.

## CLADDING DAMAGE

Film boiling operation results in high cladding temperatures within the film boiling zone. The typical appearance of a rod operated in film boiling is shown in Figure 2. As a result of the loss of cladding strength at the high temperatures and the differential between the system pressure and the rod internal pressure (typically 7 MPa), the cladding collapses onto the fuel and flows into irregularities in the fuel column. The reduction in diameter within the film boiling zone is usually fairly uniform, as shown in Figure 3. Cladding collapse into pellet interfaces (termed waisting) is shown in Figure 4. The onset of uniform cladding collapse and waisting has been found to coincide with the recrystallization process in cold worked, stress relieved zircaloy, which occurs at temperatures of about 920 K [4]. Cladding having prior irradiation (up to  $1.4 \times 10^{21}$  neutron/cm<sup>2</sup>) behaved similarly to unirradiated cladding [5]. The irradiation damage is apparently annealed out during the temperature transient prior to cladding collapse. The cladding at the elevated temperatures has sufficient ductility to accommodate the collapse and waisting strains and no failures have been detected even when up to 50% wall thinning has occurred.

The cladding collapse deformation phenomena have been investigated in out-of-pile tests by Olsen [6] as a function of pressure differential and cladding temperatures. Three deformation modes have been identified: two-point buckling, uniform collapse, and uniform collapse with waisting. The out-of-pile results are in general agreement with the PBF in-pile results; however, direct comparisons are difficult because of differences in exposure times.

As a result of the high cladding temperatures, the cladding reacts with the coolant to produce the characteristic oxide and oxygen-stabilized alpha layers at the outer surface of the cladding, as shown in Figure 5. These reaction layers are brittle and degrade the ductility of the cladding, leading to fuel rod failures as will be discussed later. The formation and extent of these reaction layers on the PBF test fuel rods appear to be consistent and

predictable by the out-of-pile, isothermal, steam-zircaloy reaction kinetics developed by Cathcart et al [7].

The collapse of the cladding onto the fuel column in the film boiling zone produces intimate contact between the cladding and the fuel. The chemical reaction which occurs between the two materials becomes significant at temperatures above 1100 K. In this reaction the  $UO_2$  fuel is reduced and oxygen diffuses into the cladding, forming an oxygen-stabilized alpha layer which embrittles the cladding in a manner similar to the cladding-coolant reaction at the outer surface of the cladding. In addition, a duplex reaction layer has often been observed between the cladding and fuel in previously unirradiated rods. Typical microstructures produced by this reaction at the inner cladding surface are also shown in Figure 5. In previously irradiated rods, only the oxygen-stabilized alpha layer has been observed [8]. A high oxygen potential may have existed in these rods as a result of oxygen or moisture pickup when the rods were opened to the atmosphere prior to testing to attach instrumentation in the plenums. This high oxygen potential may suppress the formation of the duplex layer during relatively short periods of film boiling.

Cronenberg and El-Genk [9] have presented a model to determine the oxygen distribution in zircaloy arising from fuel-cladding contact. The fuel-cladding interaction process is assumed to be diffusion-controlled and governed solely by the oxygen concentration gradient. A comparison between the analytical results of Cronenberg and El-Genk, the PBF in-pile results, and previous investigations [10, 11, 12] is shown in Figure 6. The bounding curves bracket most of the in-pile data points, indicating that a diffusional assessment of the fuel-cladding interaction appears to be appropriate, in that theory compares well with the in-pile experimental data for both unirradiated and irradiated fuel rods.

Seiffert and Hobbins [13] have analyzed the PBF results to determine the effects of hydrogen pickup on cladding microstructure in the film boiling zone. A small amount of hydrogen pickup occurred in the film boiling zones of rods which were intact prior to and during film boiling operations. Hydrides were generally dispersed in the cladding wall and no adverse effects on cladding ductility or fuel rod behavior were detected. However, three rods which were operated in film boiling with failed cladding were embrittled at room temperature to a greater extent than intact rods oxidized under similar conditions. This embrittlement appears to be associated with enhanced hydrogen absorption arising from stagnant steam conditions inside the failed rods, and with hydrogen modification of the prior-beta material as shown in Figure 7. Hydrogen concentrations as low as 300 ppm in cladding with this type of microstructure appear to significantly affect the room temperature ductility of both previously unirradiated and irradiated cladding.

### FUEL DAMAGE

Following DNB, fuel temperatures within the film boiling zone rise quickly. These high temperatures result in changes in the fuel which can affect fuel rod behavior. For high-powered rods ( $> 50$  kW/m), fuel melting frequently occurs at the center of the pellets. Some of these changes and the behavior of molten fuel during film boiling operation are discussed below.

Grain boundary separation has been observed in fuel from the film boiling zone of both unirradiated and irradiated fuel rods, a typical example of which is shown in Figure 8. The shattered fuel is in a powdered condition and has sometimes washed out of failed fuel rods. Cronenberg and Yackle [14] have examined this phenomenon and attribute it to the fuel temperature being above the equicohesive temperature of about 1900 K, at which grain boundary strength is less than the grain strength in  $UO_2$ . The calculated thermal stresses during the quench from film boiling operation are considerably higher than the grain boundary strength; thus, fracture occurs along the grain boundaries. The radial extent of the region of grain boundary fracture increases with increasing temperatures above 1900 K.

Fuel rod swelling has been observed in the film boiling zones of both previously unirradiated and, as shown in Figure 9, previously irradiated fuel rods. In unirradiated rods the swelling was caused by pellet thermal expansion and pellet volume expansion due to fuel melting. The swelling in irradiated rods was generally greater (3 to 4%), compared with unirradiated rods (< 1%). Mehner et al [15] have analyzed this phenomenon and conclude that only about half of the swelling is due to pellet thermal expansion and pellet volume expansion due to fuel melting. The remaining expansion is attributed primarily to fission gas effects. A small part of the expansion was attributed to fission-gas bubble migration to grain boundaries. However, the major contribution was concluded to be caused by fission gas bubbles trapped in the molten fuel. The molten fuel appears to act as an effective containment for the gases, sealing cracks and preventing escape of the gases to the rod plenum. The fission gas bubbles expand due to changes in surface tension and agglomeration, thereby pressurizing the molten fuel core. As bubbles continue to agglomerate, significant hydrostatic pressures may build up and cause fuel rod swelling and molten-fuel extrusion as shown in Figure 10. These fission gas effects produced no significant adverse behavior in test rods with burnups up to 17 000 MWd/t.

Rods have been operated in film boiling with up to 80% of the pellet radius molten. The molten fuel is generally contained at the center of the pellet; however, molten fuel extrusion into pellet interfaces, gaps in the fuel column (Figure 11) and radial cracks in the outer layer of the fuel pellet (Figure 10) has been observed. El-Genk [16] has modeled the case of molten fuel extrusion into a radial crack in a fuel pellet. The extent of extrusion was concluded to depend primarily on the pressure difference between the central molten zone and the fuel-cladding gap, the length and width of the crack, the molten fuel temperature, and the crack surface temperature.

#### FUEL ROD FAILURE

In the PBF tests two previously failed fuel rods were operated in film boiling, two rods failed during film boiling operation, and nine rods fractured following film boiling operation. The behavior of failed fuel rods during film boiling and rod failure mechanisms as a result of film boiling operation are discussed in this section.

Croucher [17] has assessed the behavior of failed fuel rods during film boiling and concluded that rods with defects similar to a hydride rupture outside the film boiling zone and pinhole-type or small axial crack-type defects behave similarly to intact fuel rods when operated for short times in film boiling. The significant adverse effects detected were fission product release from the failed rods and enhanced pickup of hydrogen by the cladding due to the stagnant steam inside the rod, which affects the cladding embrittlement at room temperature.

Cook [18] has concluded that a fuel rod, operated in film boiling until failure occurred due to cladding oxidation, did not fail until the cladding wall was totally oxidized to  $ZrO_2$  and oxygen-stabilized alpha with an equivalent cladding reacted value of about 24%. Preliminary results from Test PCM-5 [19] also indicate a similar failure mode in the nine-rod assembly that was operated in film boiling. These results would indicate that, at power, embrittlement failures may be predictable by a nil-ductility concept proposed by Hobson and Rittenhouse [20].

Seiffert and Hobbins [13] and Hobbins et al [21] have assessed the effects of oxygen embrittlement on rod failure during the quench from film boiling operation and during posttest handling in transport and in the hot cells. Rod failure due to oxygen embrittlement was found to be consistent with room temperature embrittlement criteria based on oxygen content in the beta zircaloy, as reported by Pawel [22], rather than embrittlement criteria based on the fractional thickness of transformed beta-phase or equivalent cladding reacted.

These results are shown in Figure 12.

The cladding on rods that failed (breached) prior to or during film boiling was embrittled to a greater extent than intact rods oxidized under similar conditions and fractured during posttest handling (see Figure 12). This additional embrittlement appeared to be associated with enhanced hydrogen absorption from the stagnant steam inside the breached rods. The hydrogen appears to affect the prior beta-phase material, resulting in room temperature embrittlement with cladding hydrogen concentrations as low as about 300 ppm.

EI-Genk [16] has assessed the conditions for melting of zircaloy cladding upon being contacted by extruded molten fuel. Cladding melting at its inner surface; i.e., potential for cladding failure upon being contacted by extruded molten fuel, depends on the temperatures of the cladding and extruded molten fuel and the metallurgical composition at the inner surface of the cladding (alpha zircaloy, beta zircaloy, or  $ZrO_2$ ). The conditions for molten fuel freezing and simultaneous cladding melting are shown in Figure 13. As would be predicted from EI-Genk's analysis, cladding melting has not occurred in the PBF tests where molten fuel has contacted the cladding. The absence of cladding melting was concluded to be due to the temperatures of the molten fuel and the cladding being below the level required to initiate cladding melting.

#### DISCUSSION AND CONCLUSIONS

The PBF tests have provided data on the primary damage mechanisms that operate on fuel rods during film boiling. Cladding damage includes deformation due to cladding collapse and waisting, oxygen embrittlement due to cladding-water and cladding-fuel reactions, and hydrogen embrittlement in failed fuel rods due to enhanced hydrogen pickup from stagnant steam conditions inside the fuel rod. Fuel damage includes grain boundary separation, molten fuel relocation, and fuel swelling. Damage mechanisms in previously unirradiated and irradiated rods are similar, except that fuel swelling due to fission gas effects occurs in irradiated fuel.

The fuel rods can operate in film boiling and incur significant damage without failure. At temperatures above 920 K, the cladding has sufficient ductility to accommodate the strains associated with cladding collapse and waisting. Failure due to oxygen embrittlement during film boiling does not occur until the cladding has been nearly completely reacted to  $ZrO_2$  and oxygen-stabilized alpha. Rod failure during quench from film boiling operation or during posttest handling, due to oxygen embrittlement, is predictable from out-of-pile isothermal steam tests. Cladding embrittlement from hydriding appears to occur at room temperature with hydrogen concentrations in the prior beta material as low as 300 ppm. Hydriding has been an embrittlement mechanism only in rods that have failed prior to or during film boiling. Molten fuel-cladding contact, with the potential for cladding melting, can occur as a result of molten fuel relocation. However, cladding melting has not occurred in the PBF tests when molten fuel has contacted the cladding, and the reaction between the cladding and fuel has not been significant. Fuel swelling has occurred in previously unirradiated rods due to thermal effects, and to a larger extent in irradiated rods due to the additional effects of retained fission gas. However, fuel swelling has not resulted in rod failure or significantly affected the behavior of rods with burnups ranging up to 17 000 MWd/t.

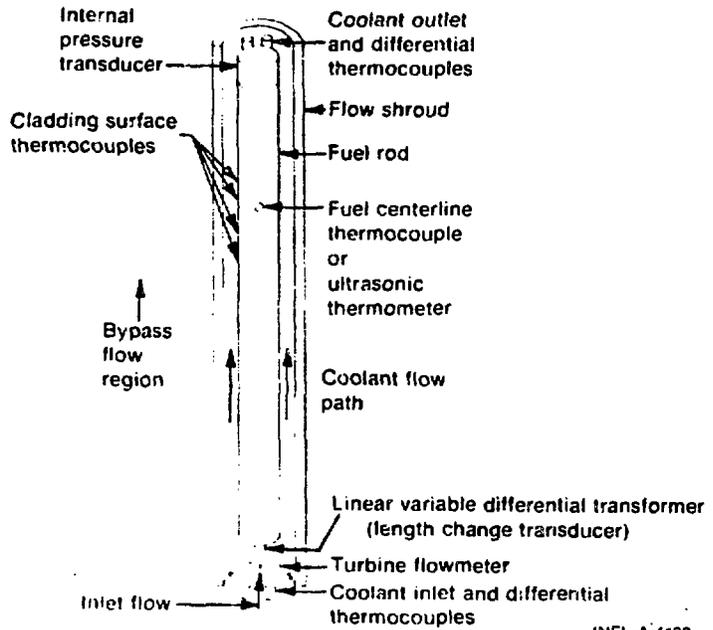
The primary rod failure mechanism in unirradiated and irradiated fuel rods is oxygen embrittlement of the cladding.

#### REFERENCES

1. A. S. Mehner et al, "Performance of Unirradiated and Irradiated PWR Fuel Rods Tested Under Power-Cooling-Mismatch Conditions," ANS Thermal Reactor Safety Meeting, Sun Valley, Idaho, July 31-August 5, 1977, Conf. 77078.

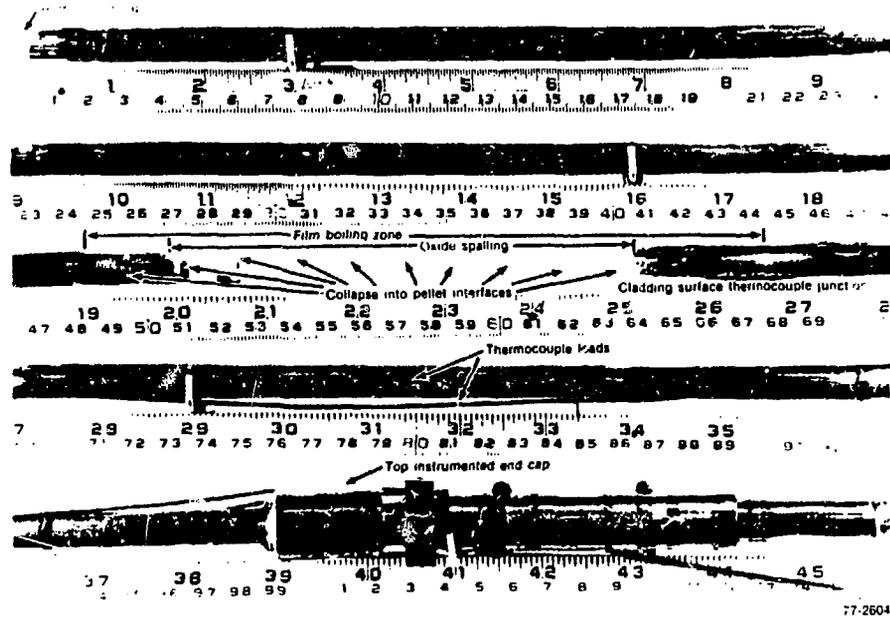
2. P. E. MacDonald et al, "Response of Unirradiated and Irradiated PWR Fuel Rods Tested Under Power-Cooling-Mismatch Conditions," Nuclear Safety, 19, No. 4 (July-August 1978) pp. 440-464.
3. W. F. Domenico et al, "Fuel Rod Failure During Film Boiling (PCM-1 Test in PBF)," Trans. Am. Nuclear Soc., 30 (November 1978) p. 406.
4. S. L. Seiffert and C. R. Smolik, "Postirradiation Examination Results for the Power-Cooling-Mismatch Test 2A," TREE-NUREG-1029 (February 1977).
5. A. S. Mehner, "Postirradiation Examination Results for the Irradiation Effects Scoping Test 2," TREE-NUREG-1022 (January 1977).
6. C. S. Olsen, "Zircaloy Cladding Collapse Under Off-Normal Temperature and Pressure Conditions," TREE-NUREG-1239 (April 1978).
7. J. V. Cathcart et al, "Zirconium Metal-Water Oxidation Kinetics IV: Reaction Rate Studies," ORNEL/NUREG-17 (August 1977).
8. A. S. Mehner and R. S. Semken, "Postirradiation Examination Results for the Irradiation Effects Test IE-1," TREE-NUREG-1194 (February 1978).
9. A. W. Cronenberg and M. S. El-Genk, "An Assessment of Oxygen Diffusion During  $UO_2$ -Zircaloy Interaction," Journal of Nuclear Materials, 78 (1978) pp. 390-407.
10. M. W. Mallett, "Solid State Reactions of Uranium Dioxide," Uranium Dioxide: Properties and Nuclear Applications, Section 7.4, J. Belle (ed.) U. S. Government Printing Office (1961) pp. 342-371.
11. L. N. Grossman and D. M. Rooney, "Interfacial Reaction Between  $UO_2$  and Zircaloy-2," GEAP-4649-ERAEC (April 1965).
12. P. Hofmann and C. Politis, "Chemical Interaction Between  $UO_2$  and Zry-4 in the Temperature Range Between 900 and 1500°C," Proceedings of the Fourth International Conference on Zirconium in the Nuclear Industry, Stratford-upon-Avon, England, June 26-29, 1978.
13. S. L. Seiffert and R. R. Hobbs, "Oxidation and Embrittlement of Zircaloy-4 Cladding from High Temperature Film Boiling Operation," NUREG/CR-0517 TREE-1327 (April 1979).
14. A. W. Cronenberg and T. R. Yackle, "An Assessment of Intergranular Fracture Within Unrestructured  $UO_2$  Fuel Due to Film Boiling Operation," NUREG/CR-0595 TREE-1330 (March 1979).
15. A. S. Mehner et al, "Fission Gas Effects in Irradiated Fuel Rods Operated in Film Boiling at High Power," Trans. Am. Nuc. Society, 28, (1978) pp. 423-424.
16. M. S. El-Genk, "An Assessment of Fuel Melting, Radial Extrusion and Cladding Thermal Failure During a Power-Cooling-Mismatch Event in Light Water Reactors," NUREG/CR-0500, TREE-1270 (to be published).
17. D. W. Croucher, "Behavior of Defective PWR Fuel Rods During Power Ramp and Film Boiling," NUREG/CR-0283, TREE-1267 (February 1979).

18. B. A. Cook, "Fuel Rod Material Behavior During Test PCM-1," NUREG/CR-0757, TREE-1333 (May 1979).
19. D. T. Sparks et al, "Film Boiling Behavior in a Nine Rod Cluster," Trans. Am. Nuc. Soc., 30 (November 1978) p. 404.
20. D. O. Hobson and P. L. Rittenhouse, "Embrittlement of Zircaloy-Clad Fuel Rods by Steam During LOCA Transients," ORNL 4758 (January 1972).
21. R. R. Hobbins et al, "The Embrittlement of Zircaloy Clad Fuel Rods Irradiated Under Film Boiling Conditions," 4th International Conference on Zirconium in the Nuclear Industry, Stratford-upon-Avon, England, June 26-29, 1978.
22. R. E. Pawel, "Oxidation Diffusion in Beta Zircaloy During Steam Oxidation," Journal of Nuclear Materials, 50 (1974) pp. 247-258.



INEL-A-4190

Fig. 1 Test configuration.



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Fig. 2 Film boiling zone on Rod IE-C20, Test IE-5, showing sharp surface color change at lower film boiling boundary, cladding collapse into pellet interfaces, and oxide spalling within the film boiling zone..

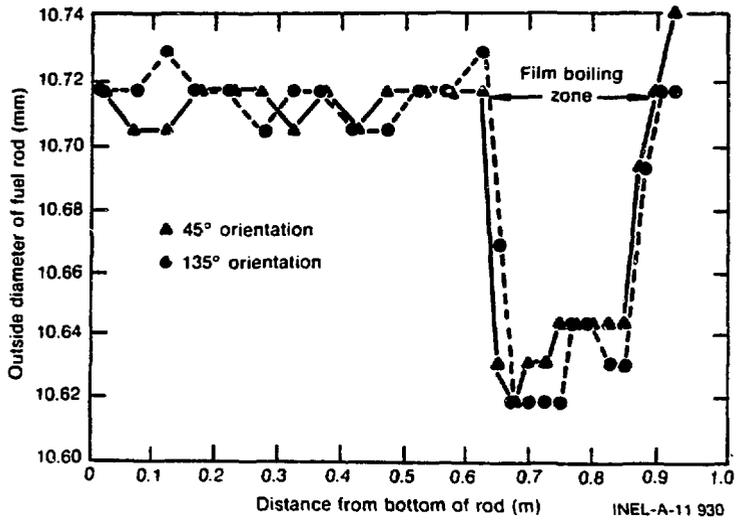


Fig. 3 Cladding outside diameter of Rod UTA-0006, Test PCM 8-1 RF, showing reduction in diameter within the film boiling zone of an irradiated fuel rod.

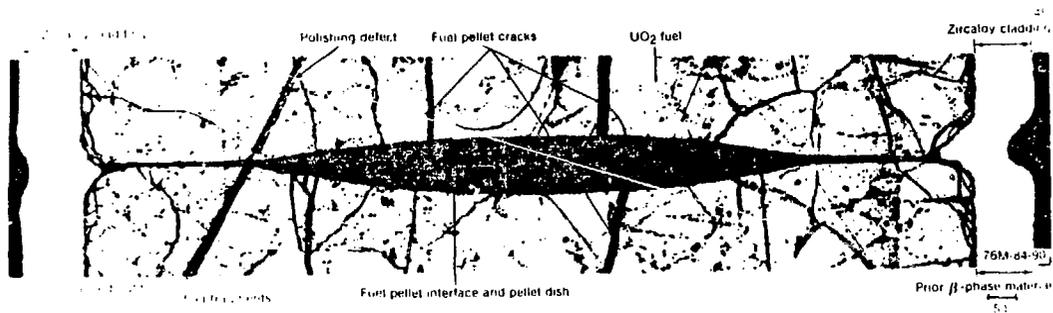


Fig. 4 Example of cladding flow into a pellet-to-pellet gap in Rod UTA-0007, Test PCM-2A.

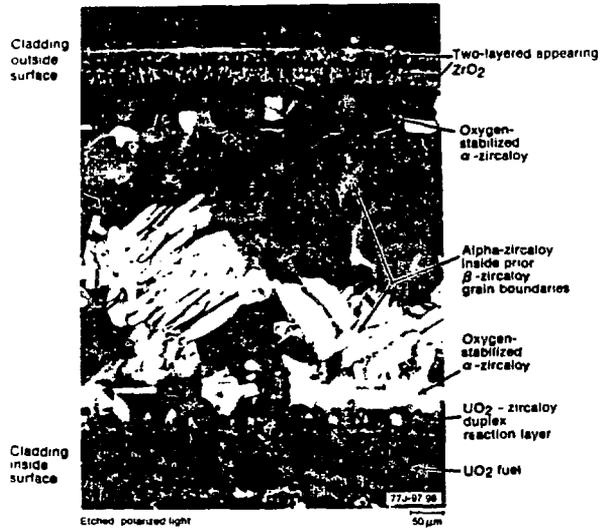


Fig. 5 Typical cladding microstructure in region of film boiling zone on Rod IE-020, Test IE-5.



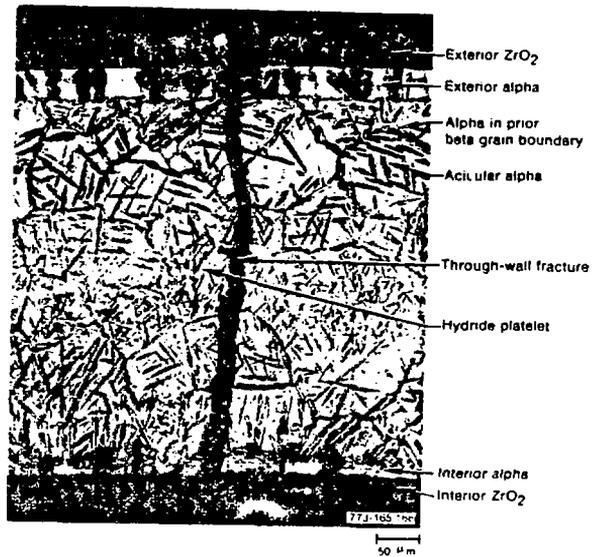


Fig. 7 Cladding microstructure from Rod IE-019, Test IE-5, showing hydride effects that result in room temperature embrittlement.

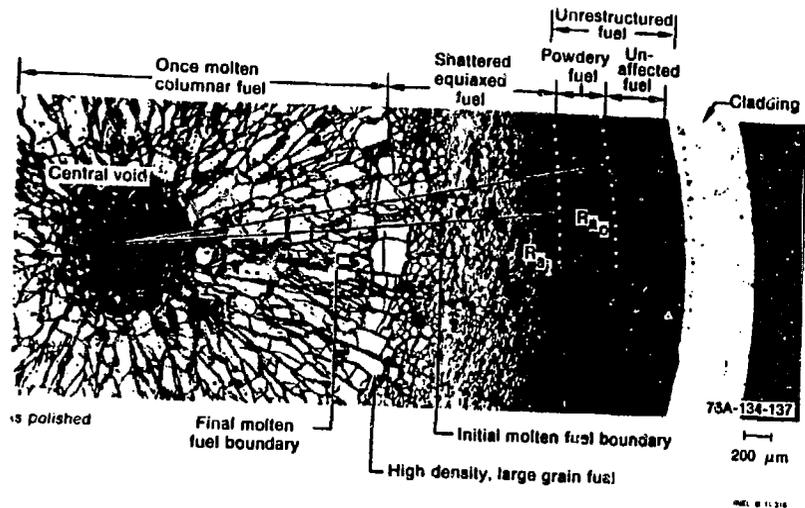


Fig. 8 Transverse section showing typical fuel microstructure and grain boundary shattering across a fuel pellet from the film boiling zone of a previously unirradiated fuel rod, Rod IE-001, IE Scoping Test 1.

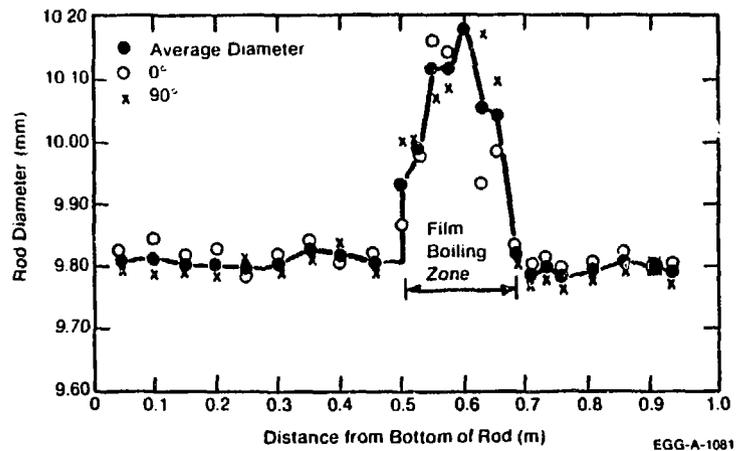


Fig. 9 Posttest diametral measurements showing diameter increase in the film boiling zone of a previously irradiated fuel rod, Rod IE-010, Test IE-1.

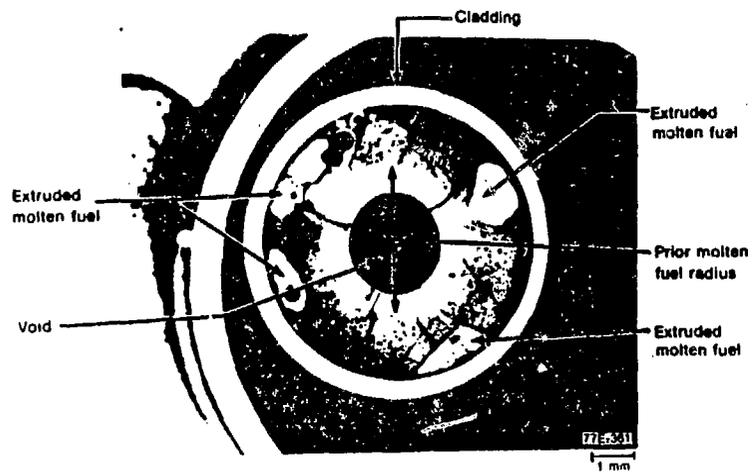


Fig. 10 Molten fuel extrusion in an irradiated fuel rod, Rod IE-016, Test IE-3.

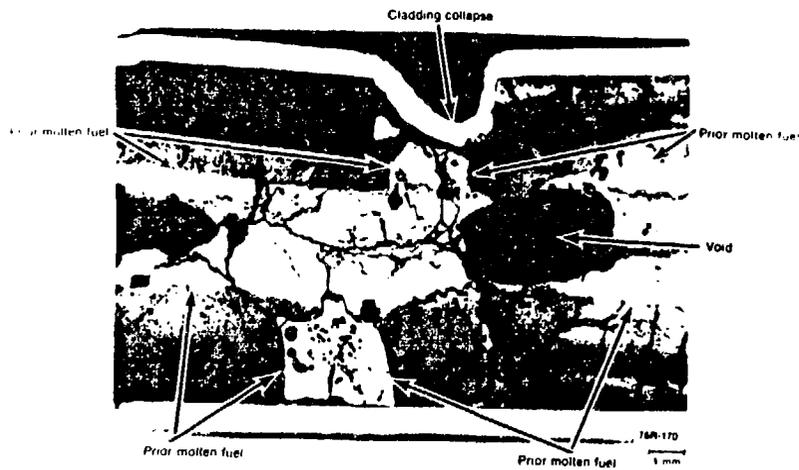


Fig. 11 Longitudinal section showing molten fuel extrusion in the film boiling zone of a previously irradiated fuel rod, Rod IE-009, Test IE-1.

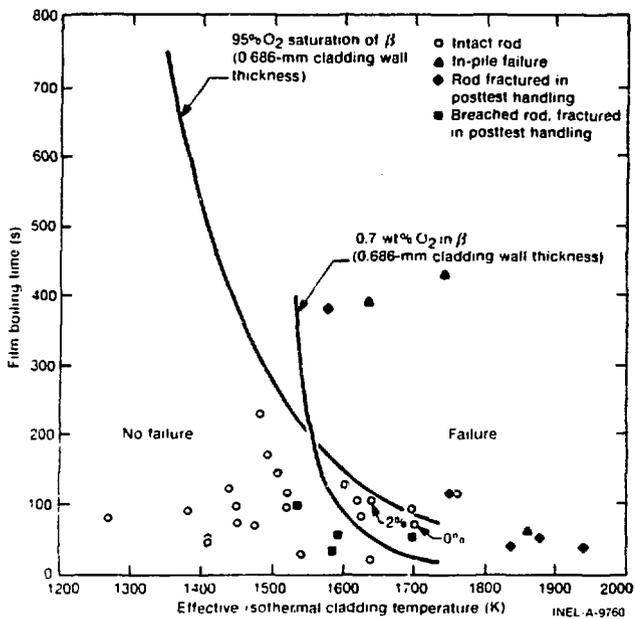


Fig. 12 Failure criteria for intact fuel rods tested under PCM conditions.

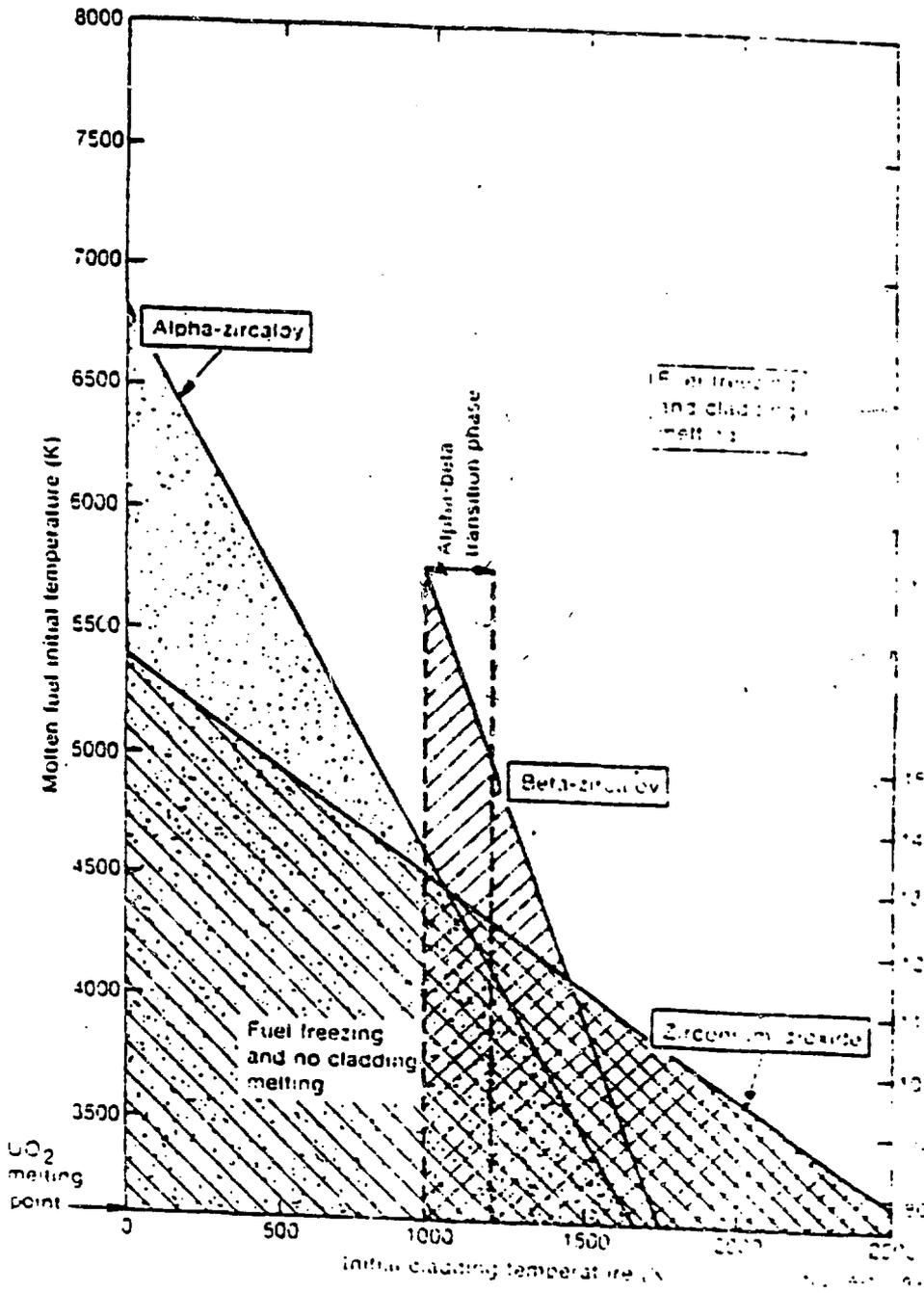


Fig. 13 Initial temperatures map for initially molten  $UO_2$  contacting initially solid zircaloy cladding.