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BEHAVIOR OF EBR-II MK-V-TYPE FUEL ELEMENTS IN SIMULATED LOSS-OF-FLOW TESTS*

Y. Y. Liu, H. Tsai, M. C. Billone, J. W. Holland,** and J. M. Kramer**

Materials and Components Technology Division
ARGONNE NATIONAL LABORATORY
Argonne, Illinois 60439-4838 USA

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**Reactor Engineering Division.

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ABSTRACT

Three furnace heating tests were conducted with irradiated, HT9-clad and U-19wt.%Pu-10wt.%Zr-alloy fuel, Mk-V-type fuel elements in the Alpha-Gamma Hot Cell Facility at Argonne National Laboratory, Illinois. In general, very significant safety margins for fuel-element cladding breaching have been demonstrated in these tests, under conditions that would envelop a bounding unlikely loss-of-flow event in EBR-II. Highlights of the test results will be given, as well as discussions of the cladding breaching mechanisms, axial fuel motion, and fuel surface liquefaction found in high-temperature testing of irradiated metallic fuel elements.

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1. Introduction

The next step in the development of metal fuels for the Integral Fast Reactor (IFR)[1] is the conversion of the core of the Experimental Breeder Reactor (EBR)-II at Argonne National Laboratory-Idaho (Argonne-West) to one containing the ternary U-20wt.%Pu-10wt.%Zr alloy fuel with HT9 cladding, designated the Mk-V core. The plutonium content, together with fission products left after recycling, makes Mk-V fuel a prototype for the Integral Fast Reactor. Mk-V fuel will be manufactured at the Fuel Cycle Facility (FCF) currently being outfitted at Argonne-West. The combination of a liquid metal reactor (e.g., EBR-II) and an integral fuel cycle facility (e.g., FCF) producing recycled fuel for the reactor embodies the IFR concept and demonstrates closure of the fuel cycle.

A safety case is being prepared for Mk-V core conversion. Design criteria have been established to ensure that the Mk-V fuel elements will meet their functional requirements (e.g., power level) and performance objectives (e.g., burnup) safely and reliably. These criteria are based on knowledge of damage mechanisms in metallic fuel elements under normal and off-normal reactor operating conditions. For example, given the current burnup goal of the Mk-V fuel, i.e., 10 at.%, irradiation-induced swelling is not considered a cladding damage mechanism because of the swelling resistance of the HT9 ferritic/martensitic steel. Creep deformation, on the other hand, is a potential cladding damage mechanism, especially during off-normal temperature excursions. Another potential cladding damage mechanism is fuel/cladding metallurgical interaction involving either solid-state interdiffusion at relatively low temperatures, or fuel liquefaction and cladding matrix dissolution at high temperatures. Both damage mechanisms have been studied in separate-effects tests with irradiated, non-fueled and fueled specimens in the Fuel Cladding Transient Tester (FCTT)[2] and the Fuel Behavior Test Apparatus (FBTA),[3] respectively. The analytical correlations, derived from the separate-effects test data and incorporated into the LIFE-METAL and FPIN-2 fuel behavior modeling codes,[4, 5] form the basis for fuel performance analysis under normal and off-normal reactor operating conditions.

Mk-V fuel performance under normal reactor operation will be validated in a surveillance program that monitors selected Mk-V fuel assemblies during EBR-II irradiation. For off-normal conditions, Mk-V fuel performance will be evaluated through a combination of hot-cell and in-reactor experiments and analysis with the FPIN-2 and LIFE-METAL codes. This paper will present results of three hot-cell furnace heating tests with irradiated U-19wt.%Pu-10wt.%Zr/HT9, Mk-V-type fuel elements. The thermal conditions of these tests were designed to envelop a bounding unlikely loss-of-flow (LOF) event, UN-1, in EBR-II, during which the calculated peak cladding temperature would reach 776°C for less than 2 minutes. The principal objectives of the tests were to (1) determine the safety margin of the fuel element, (2) investigate cladding breaching behavior, and (3) provide data for validation of the FPIN-2 and LIFE-METAL codes.

2. Experimental

The three tests, designated FM-2, FM-4 and FM-5, were conducted in the Whole-Pin Furnace (WPF) system located in the Alpha-Gamma Hot-Cell Facility (AGHCF) at Argonne National Laboratory-Illinois (Argonne-East). Detailed description of the WPF system and its performance characteristics can be found in Ref. 6. Figure 1 is a schematic of the WPF system showing a stainless steel test capsule containing an irradiated fuel element. The capsule is normally evacuated and sealed during the test. Two pressure transducers at the top of the capsule provide the means for cladding breach detection, i.e., by measuring the pressure rise due to fission-gas release intc the sealed system. The temperature along the length of the fuel element is measured by six sheathed, chromel-alumel thermocouples located at different elevations in the annulus between the fuel element and the capsule. A ceramic insulated, bare-wire Pt/Pt-10%Rh control thermocouple, which is welded on the outside of the capsule, serves to maintain the desired test temperatures. Outside the capsule is a quartz tube that provides a channel for helium cover gas flow, which provides an inert atmosphere for the capsule during the test.

The furnace is a radiant heating chamber powered by four longitudinal infrared-filament lamps. Elliptically shaped aluminum reflectors behind each lamp provide highly efficient focusing of the radiant energy onto the centerline of the furnace, where the test fuel element is placed. Control of the furnace is based on output of the control thermocouple via a feedback algorithm in a microcomputer. Typical accuracy in an extended heating cycle (up to 36 h) is within ±2°C of the target temperature. Because the furnace provides external heating of the fuel element, the fuel temperature profile is flat under steady-state heating conditions. For metal fuels with thermal conductivities comparable to that of cladding steels, the fuel temperature profile in loss-of-flow accidents at decay heat levels would be nearly flat, and, hence, closely simulated by external heating.

The fuel elements used in the furnace tests were irradiated in EBR-II under steady-state conditions. For each fuel element, pre- and posttest examination typically included gamma scanning and/or neutron radiography to determine change in the fuel column length, measurement of element diameter to determine cladding deformation, and metallography to determine fuel/cladding metallurgical interaction and microstructure. The fission gas released from the breached fuel element was also collected in the fission-gas measurement system for mass spectrometric analysis.

3. Test Description and Results

Table 1 summarizes the Mk-V fuel parameters and key variables in the three FM tests. Compared to Mk-V fuel, the slightly lower Pu content in the test fuel, i.e., 19 vs. 20 wt.%, would be insignificant from the viewpoint of fuel/cladding metallurgical interaction.[3] Differences in the other fuel-element parameters, e.g., cladding thickness, plenum-to-fuel volume ratio and burnup, led to tests that would yield conservative estimates of the safety margin for the Mk-V fuel. The test parameters, e.g., temperature and duration, too, were more aggressive than the off-normal conditions of the unlikely LOF event, and, thus were conservative from the standpoint of Mk-V element survivability.

3.1 Run-to-cladding-breach tests (FM-2 and FM-4)

The FM-2 and FM-4 tests were run-to-cladding-breach tests conducted at constant temperatures, 820 and 770°C, respectively. The measured cladding breaching times were 112 min. for the low-burnup FM-2 fuel element and 68 min. for the high-burnup FM-4 fuel element. Figure 2 shows the recorded temperatures in the two tests, along with the calculated cladding temperature history in the Mk-V fuel during the bounding UN-1 LOF event in EBR-II. Compared to the total duration of ≤2 min. of the LOF event, both FM-2 and FM-4 tests have demonstrated very significant safety margins for cladding breaching for the U-19wt.%Pu-10wt.%Zr/HT9, Mk-V-type fuel elements. The shorter cladding breaching time in FM-4 at a lower test temperature than FM-2 was due to the higher fission-gas pressure in the high-burnup FM-4 fuel element, which also resulted in a different type of cladding breach from that of the low-burnup FM-2 element (see below).

The FM-2 fuel element cladding breached near the fuel top in the form of a small axial crack with a length of ~2.0 mm and an opening of 0.9 mm at the widest point. A typical transverse section near the mid-height of this crack is shown in Fig. 3. Fuel/cladding metallurgical interaction, which reduced the cladding to a thin membrane near the crack tip, was prominent along the entire length of the crack. This localized wall thinning apparently played a dominant role in cladding breaching, with fission-gas pressure causing the final rupture of the thin membrane. However, the rupture was benign as indicated by the crack dimension and the relatively small diametral strains (2.3 to 4.3%) near the crack tip. In contrast to the benign cladding breach in the fuel column region of the low-burnup FM-2 fuel element, the high-burnup FM-4 fuel element cladding breached in the plenum region, at ~150 mm above the fuel top, with significant cladding deformation (up to ~15% strain close to the breach). The breach, shown in Fig. 4 with a length of 12.5 mm and a width of 3.5 mm, was a rupture much larger than the crack in FM-2. Examination of the breached cladding section showed no remnants of fuel, nor any evidence of fuel/cladding metallurgical interaction. The breach was a classical creep rupture of the HT9 cladding caused by fission-gas pressure loading. Apparently all of the plenum fission-gas inventory was collected from the breached element, i.e., 78% vs. 77% measured for a steady-state sibling fuel element.

Extensive fuel/cladding metallurgical interaction occurred in the low-burnup FM-2 fuel element, causing not only cladding interaction, but also fuel liquefaction, foaming and axial fuel motion which measured ~51 mm above the as-irradiated fuel top. An example of foamed fuel that filled the entire fuel cross section at the fuel top has been shown in Fig. 3. Another example is shown in Fig. 5 for two adjacent longitudinal sections, between X/L = 0.76 and 0.85, where the morphology consisted of foamed fuel filling the fuel cross section, large central cavities, and isolated pores surrounded by rather compact, liquefied fuel with much fewer porosities. Some of the closed pores apparently trapped fission gases, since only ~12% of the expected plenum fission-gas inventory was collected from the breached FM-2 fuel element. (Indirect evidence of fission-gas trapping was also indicated when the hot cell stack monitor registered a sharp increase in ^{85}Kr activity shortly after fuel-element sectioning.) As the temperature became lower toward the bottom of the fuel column, the areal fuel liquefaction decreased from 100% at X/L = 0.68 to ~11% at X/L = 0.54, and then completely disappeared at X/L= 0.48 where the temperature was ~760°C during the test.

Far less fuel/cladding metallurgical interaction occurred in the high-burnup FM-4 fuel element that was tested at a lower temperature with a shorter duration than those of the FM-2 fuel element. Figure 6 shows ~20% areal fuel liquefaction at X/L = 0.96, where the temperature was 750°C during the test. (Maximum cladding penetration at this location was ~51 μm, or 13.3% of the original cladding thickness.) The limited fuel liquefaction and foaming resulted only in a modest growth of the FM-4 fuel column, i.e., ~3 mm based on posttest neutron radiography.

3.2 Proof test (FM-5)

The FM-5 test was a proof test conducted to demonstrate that a high-burnup, U-19wt.%Pu-10wt.%Zr/HT9, Mk-V-type fuel element can survive a simulated unlikely LOF event in EBR-II with only minor incremental cladding deformation and wastage and no cladding breach. The thermal conditions of the test were designed to envelop the cladding temperature history in the UN-1 LOF event with a peak instantaneous temperature of 776°C. (The actual temperature profile in the FM-5 test can be found in the paper by Kramer, et al., this Symposium.)

The high-burnup FM-5 fuel element endured the simulated UN-1 temperature transient without cladding breaching and with negligible incremental cladding deformation. Based on the measured thermocouple temperatures, a thermal analysis has established that the top portion ($0.86 \leq X/L \leq 1.0$) of the fuel element was heated above 660°C for a period of ~40 s during the test. Posttest destructive examination of the fuel element sections that experienced the highest temperature showed no evidence of any fuel at the surface being above a solidus temperature (Fig. 7). Also, the measured cladding wastages were comparable to that of the steady-state sibling fuels, indicating very little incremental cladding interaction during the test.

4. Discussion

4.1. Cladding breaching mechanisms

For the low-burnup FM-2 fuel element that breached near the top of the fuel after 112 min. at 820°C, the breaching mechanism was apparently localized cladding wall thinning due to fuel/cladding metallurgical interaction, in conjunction with fission-gas pressure loading. The FBTA tests conducted with irradiated metal fuels over the last few years have identified three types of fuel/cladding metallurgical interactions, i.e., solid-state diffusional interaction, grain boundary liquid-phase penetration, and matrix dissolution, depending on test temperatures, fuel burnup, and cladding types.[3] For low-burnup fuels with HT9 cladding in the temperature range of the FM-2 test, cladding interaction in the form of matrix dissolution prevailed. This was indeed the case for the FM-2 fuel element, as shown in Fig. 8, which compares matrix dissolution in a sibling HT9 cladding after a 1-h, 780°C FBTA test with that in the FM-2 cladding at the same temperature. The latter had a deeper interaction because of the longer duration (112 min.) at 780°C.

Based on extrapolation of the FBTA data on low-burnup sibling fuels, matrix dissolution would require 235 min. for complete penetration of the FM-2 cladding. Without cladding dissolution and considering gas pressure loading only, the predicted cladding stress-rupture time for the FM-2 fuel element is 455 min. at a plenum temperature of 820°C. The experimental breaching time of 112 min., which is significantly shorter than either of the above failure times, indicates a strong synergism between the two potential cladding breaching mechanisms, i.e., fuel/cladding metallurgical interaction and fission-gas pressure loading.

For the high-burnup FM-4 fuel element that breached in the plenum after 68 min. at 770°C, the breaching mechanism was apparently creep rupture of the HT9 cladding caused by fission-gas pressure loading. Fuel/cladding metallurgical interaction was not a factor in the breaching of the FM-4 element. Contrary to the reasonably close pretest predictions made by the LIFE-METAL and FPIN-2 codes for the low-burnup FM-2 fuel element (see the paper by Kramer, et al., this Symposium), both codes significantly underpredicted the cladding breaching time for the high-burnup FM-4 fuel element. One reason for the underprediction could be that the current HT9 correlations used in the codes do not account for microstructural changes, such as grain coarsening and secondary carbide precipitation, and the ensuing strengthening of the ferritic/martensitic steel during high-temperature operation.[7] This issue is currently being addressed at Argonne-East in an experimental program on high-temperature mechanical properties of the HT9 cladding.

4.2 Axial fuel motion

Upward fuel motion inside the cladding of ~51 and 3 mm occurred during the FM-2 and FM-4 tests, respectively. The mechanism for upward fuel motion is apparently fuel foaming, made possible by the fisson-gas inventory in the fuel that is undergoing solid/liquid phase transformation, i.e., liquefaction. Foaming and gross swelling has been observed in several irradiated nuclear fuel systems (e.g., uranium metal,[8] UO_2/Zircaloy under a reducing environment,[9] U-Al fuel alloy[10]) during melting or formation of low-temperature liquid/solid phases. In the FM-2 and FM-4 fuel elements, fuel liquefaction resulted from diffusion of Fe from the HT9 cladding into the U-19wt.%Pu-10wt.%Zr fuel, which lowered the solidus temperature of the fuel. (Fission products lanthanides at fuel/cladding interaface probably played a role in fuel surface liquefaction in the high-burnup FM-4 fuel, but was not a factor in the low-burnup FM-2 fuel, see below.) As the fuel became liquefied, the mobility of fission gas atoms in the fuel increased and small fission-gas bubbles coalesced into larger ones. Because of the cladding restraint, however, expansion of the foamed fuel was directed upward into the plenum region. Similar foam expansion inside the cladding was also observed in the metal fuel elements tested in the TREAT reactor, where fuel melting occurred under much faster (≤8 s) transient overpower coniditions.[11] The upward expansion of liquefied fuel, not slumping, inside the cladding is highly desirable from the viewpoint of reactor safety.

4.3 Fuel surface liquefaction

For metallic fuel elements in general, and Mk-V fuel elements in particular, fuel surface liquefaction at fuel/cladding interfaces can occur at temperatures much lower than the inherent melting point of the fuel. In the case of MK-V fuel elements, the agents responsible for fuel surface liquefaction involve principally Fe diffused from the HT9 cladding, fuel alloy constituents, U and Pu, and fission products lanthanides, e.g., Ce, Pr, and Nd. The lanthanides, which become more abundant with burnup, are important to fuel surface liquefaction because they have relatively low eutectic temperatures with Fe.

Fuel surface liquefaction in the low-burnup FM-2 fuel element was found terminated at X/L = 0.48, where the temperature was ~760°C during the 112-min. test. In the high-burnup FM-4 fuel element, fuel surface liquefaction was found terminated at X/L = 0.51, where the temperature was 675°C during the 68-min. test. These fuel liquefaction threshold temperatures apparently follow the same pattern found in FBTA tests on the sibling fuels. For example, recent FBTA tests with high-burnup (11 at.%), U-19wt.%Pu-10wt.%Zr/HT9 fuels have shown a temperature threshold for fuel surface liquefaction between 660 and 675°C, which was ~75°C lower than the previously reported temperature threshold for lower burnup fuels. (Fuel surface liquefaction in high-burnup sibling fuels was found after 1 h at 675°C and 12 h at 660°C, but not after 36 h at 650°C.) Deeper penetration of lanthanides into the HT9 cladding was also noted in the higher burnup FBTA specimens (see the paper by Cohen et al., this Syposium). Unlike fuel melting in oxide fuel elements which has traditionally been equated with fuel element failure, fuel surface liquefaction in metal fuel elements should not be regarded as failure, particularly when fuel/cladding interface temperature only momentarily exceeds the liquefaction temperature threshold based on FBTA tests with a minimum 1-h test duration. The high-burnup fuel element in the FM-5 test provided a good example: Not only did the fuel element maintain its integrity, there was no evidence of any fuel surface liquefaction, even though the fuel/cladding interface temperature was above 660°C for a brief period during the test.

5. Conclusions

1. Significant temporal safety margins for cladding breaching, up to one hour or more, have been demonstrated in two run-to-cladding-breach tests with irradiated U-19wt.%Pu-10wt.%Zr/HT9, Mk-V-type fuel elements. A proof test has also demonstrated that a high-burnup, U-19wt.%Pu-10wt.%Zr/HT9 fuel element can endure a simulated bounding unlikely LOF event in EBR-II with negligible damage to fuel and cladding.

2. Two cladding breaching mechanisms were operative in the FM-2 and FM-4 fuel elements. For the low-burnup FM-2 fuel element that failed near the fuel top after 112 min. at 820°C, the breaching mechanism was cladding thinning due to fuel/cladding metallurgical interaction, in conjunction with fission-gas pressure loading. For the high-brunup FM-4 fuel element that failed in the plenum after 68 min. at 770°C, the breaching mechanism was creep rupture of the HT9 cladding caused by fission-gas pressure loading.

3. Fuel/cladding metallurgical interaction is responsible for cladding matrix dissolution, fuel liquefaction, foaming and axial fuel motion found in high-temperature testing of irradiated metallic fuel elements. Whereas matrix dissolution of the cladding reduces its load-bearing

capability and accelerates breaching, foaming upward fuel motion is generally desirable from the viewpoint of reactor safety.

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Table 1. Summary of Mk-V Fuel Parameters and FM Test Variables

Fuel Element	Fuel/Cladding Materials	Cladding Thickness (mm)	Plenum/Fuel Vol. Ratio	Peak Fuel Burnup (at.%)	Test Temp. (°C)	Test Duration (min)
Mk-V	U-20Pu-10Zr/HT9	0.457	1.5	10.0	776[a]	<2[a]
FM-2	U-19Pu-10Zr/HT9	0.381	1.0	3.0	820	112[b]
FM-4	U-19Pu-10Zr/HT9	0.381	1.5	11.4	770	68[b]
FM-5	U-19Pu-10Zr/HT9	0.381	1.5	11.4	780	3

[a]The temperature and duration are those of the Mk-V fuel undergoing a bounding UN-1 unlikely loss-of-flow event in EBR-II.

[b]Cladding breaching times during temperature hold at 820 and 770°C, respectively.

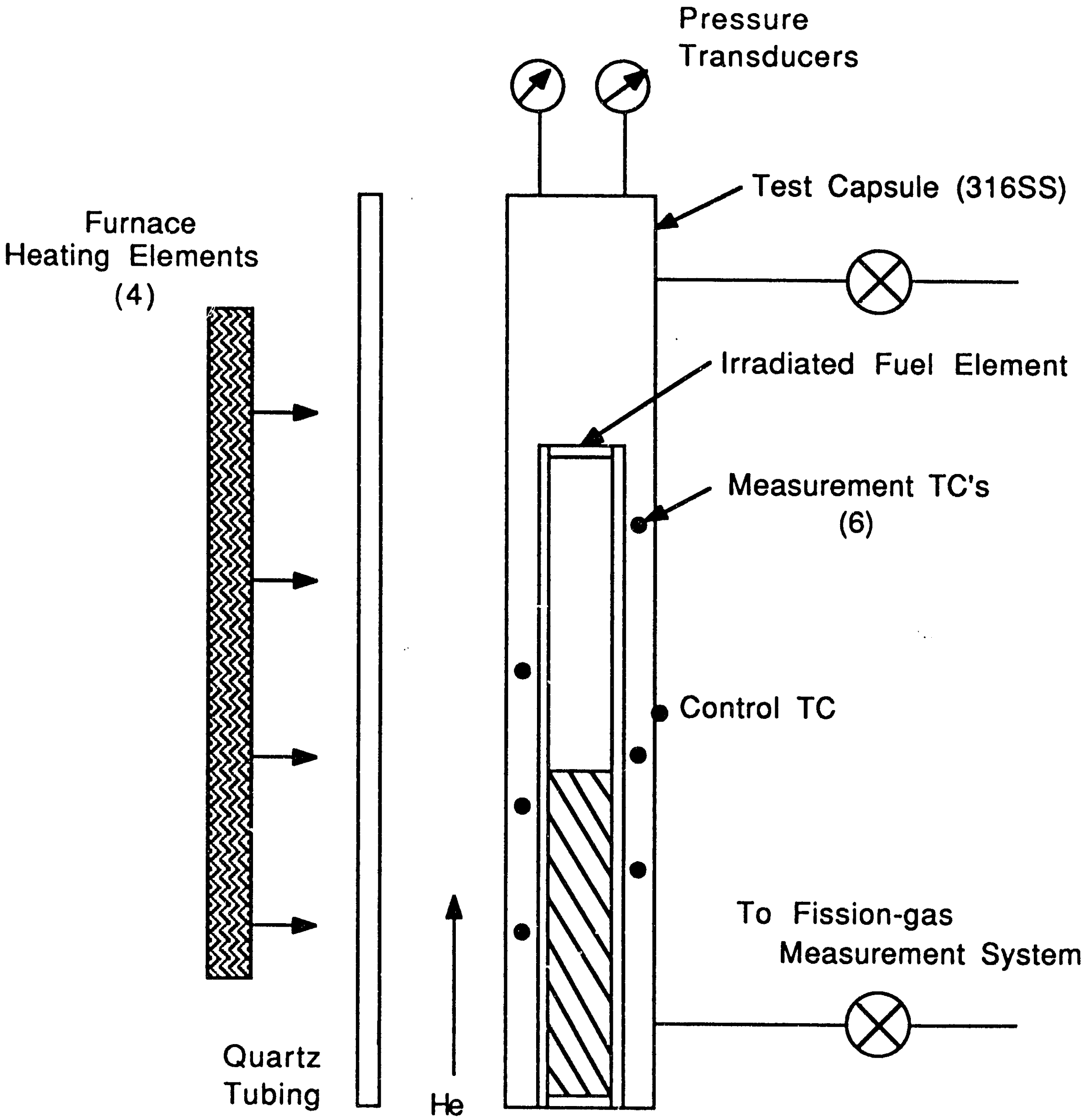


Fig. 1. Schematic of the Whole-Pin Furnace System.

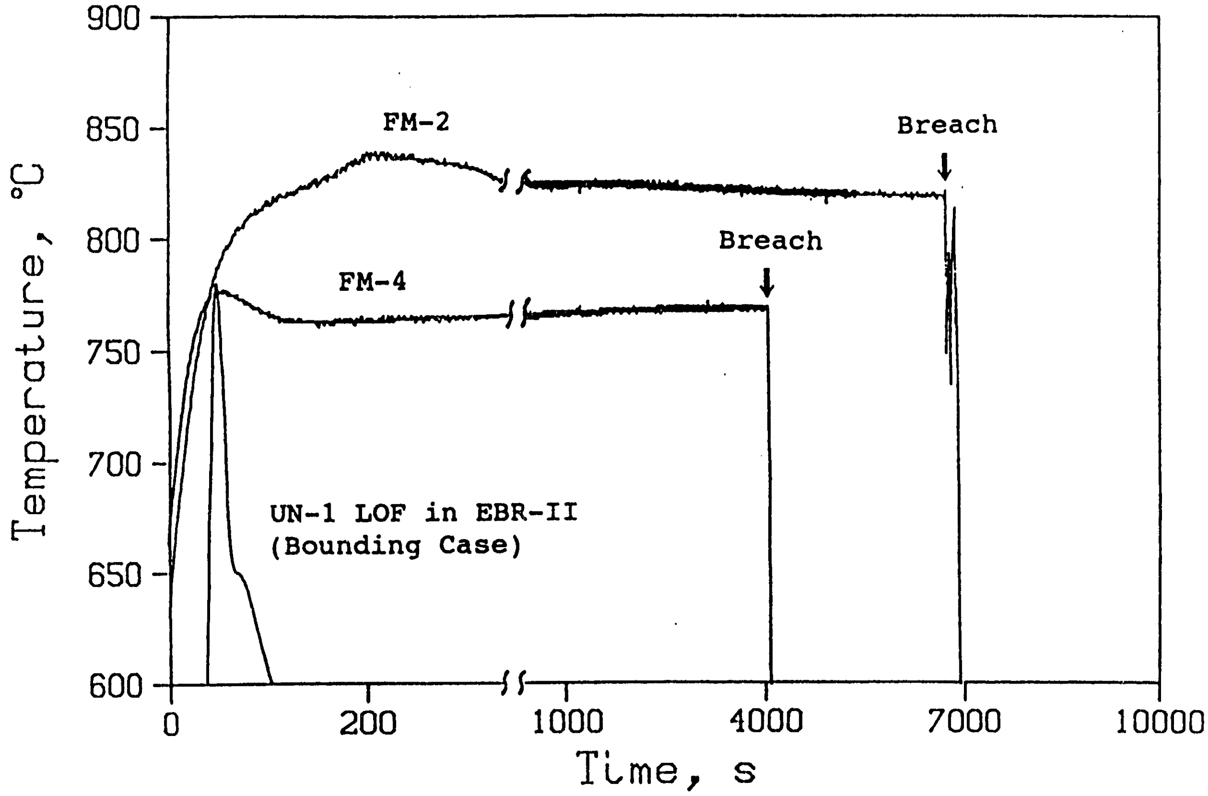


Fig. 2. Comparison of Cladding Breaching Margin (Time) in High-burnup (11.4 at.%) FM-4 Test and Low-burnup (2.9 at.%) FM-2 Test.

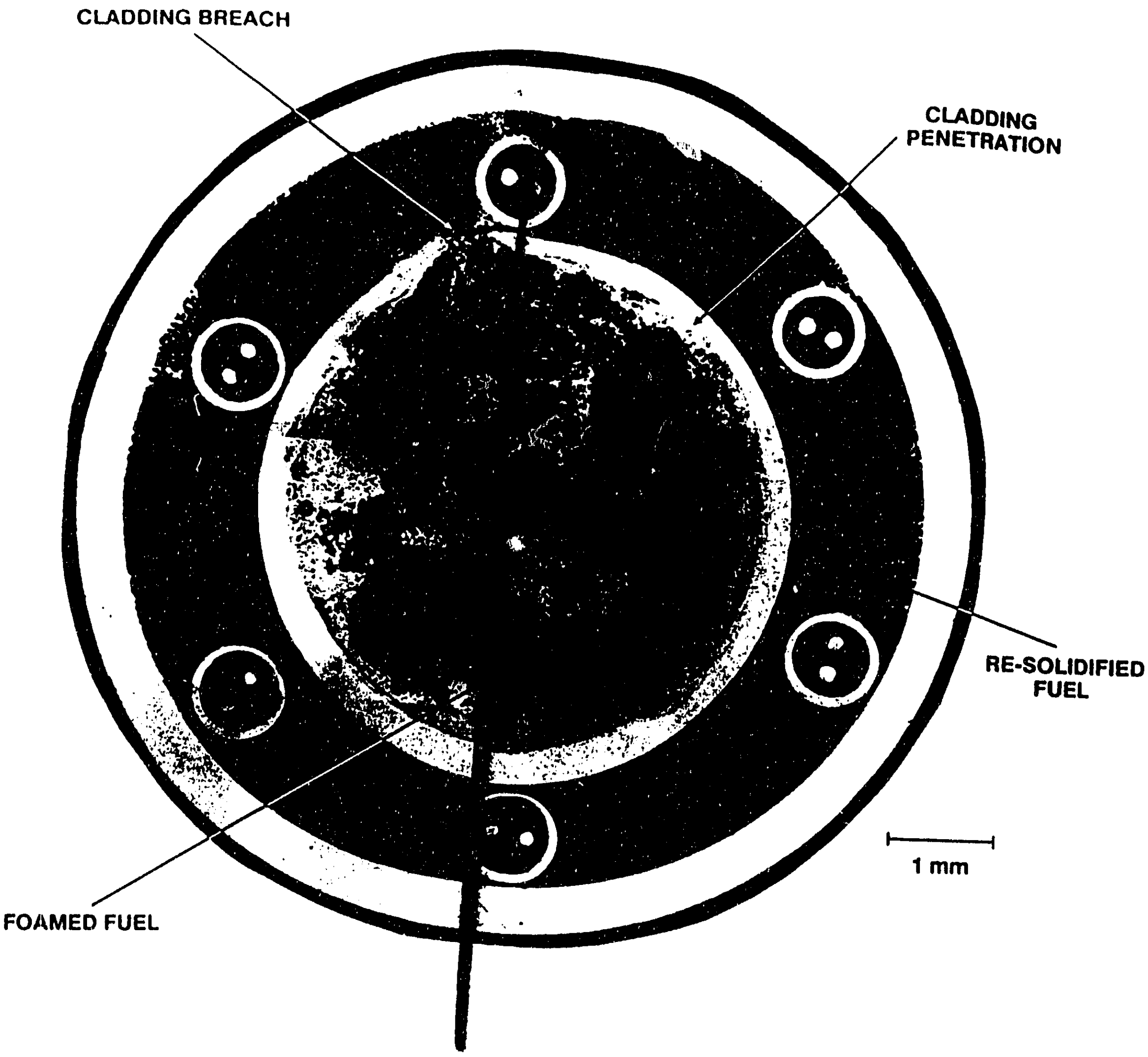


Fig. 3. Cladding Breach (Near Fue Top) in the Low-burnup, Mk-V-Type Fuel Element in the FM-2 Test.

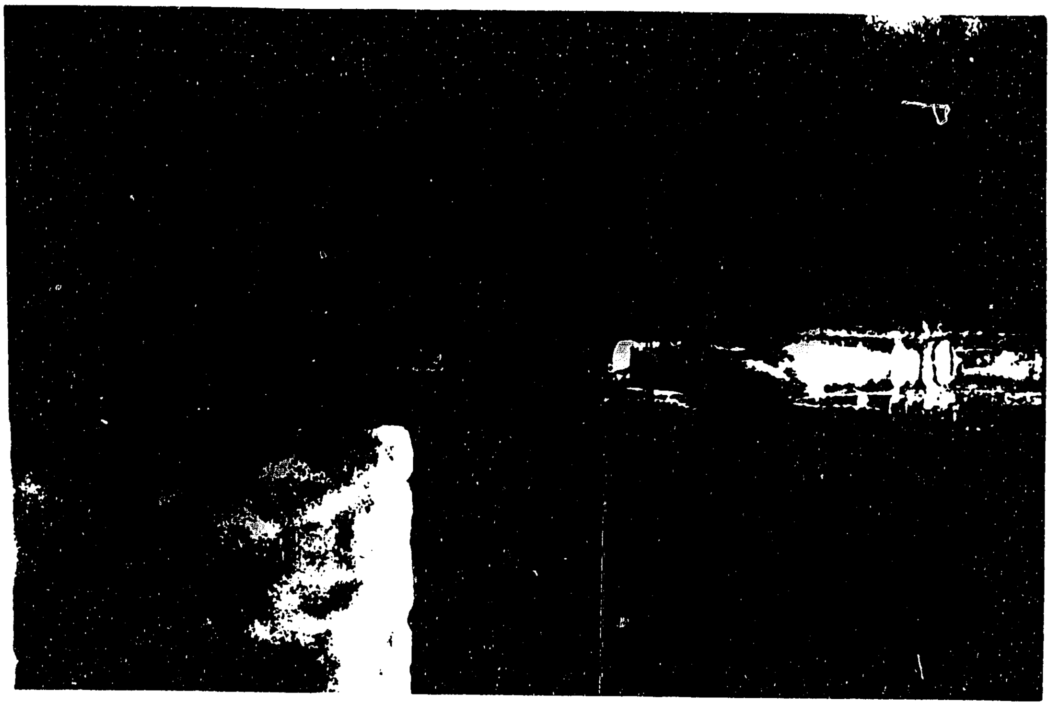


Fig. 4. Cladding Breach in the Plenum Region of the High-burnup, Mk-V-Type Fuel Element in the FM-4 Test. Fuel element top is to the right, 2X.

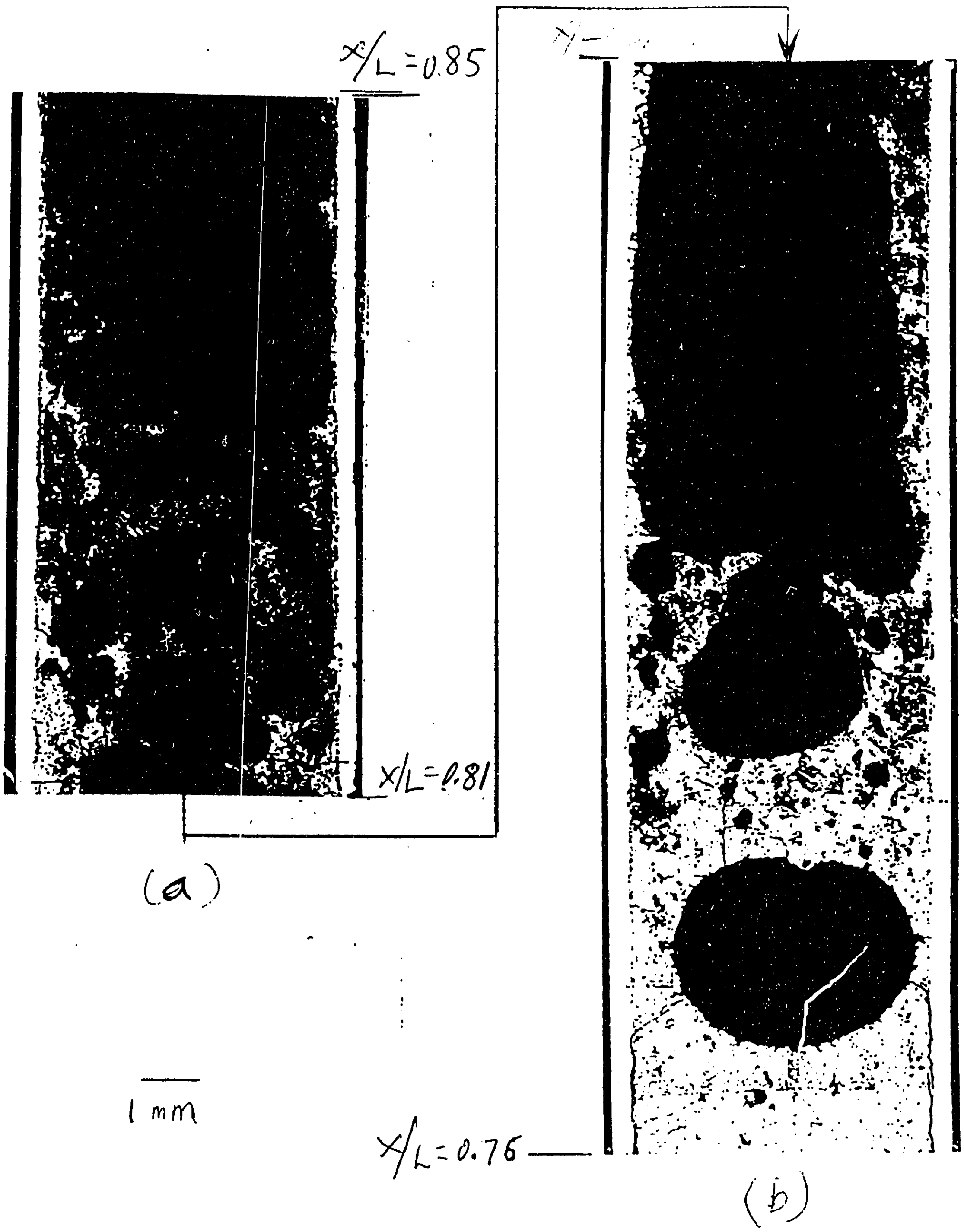


Fig. 5. Longitudinal Sections of the FM-2 Fuel Element Between X/L = 0.76 and 0.85, Showing Foamed Fuel Between Two Central Cavities in (a) Isolated Pore and (b) the Surrounding Relatively Pore-free, Resolidified Fuel.

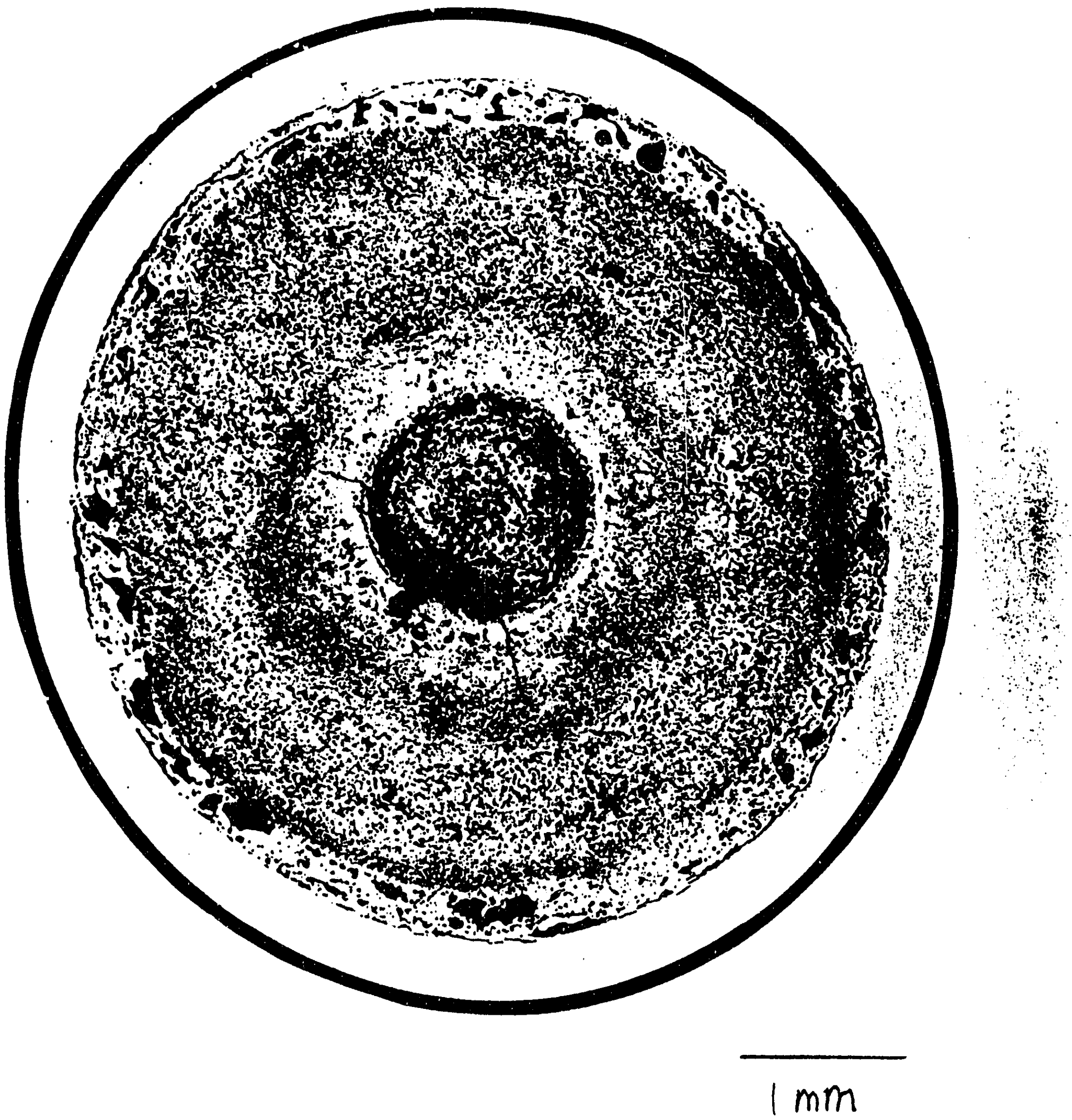


Fig. 6. Fuel Surface Liquefaction in the High-burnup FM-4 Fuel Element at X/L = 0.96. This location was heated at 750°C for 68 min.

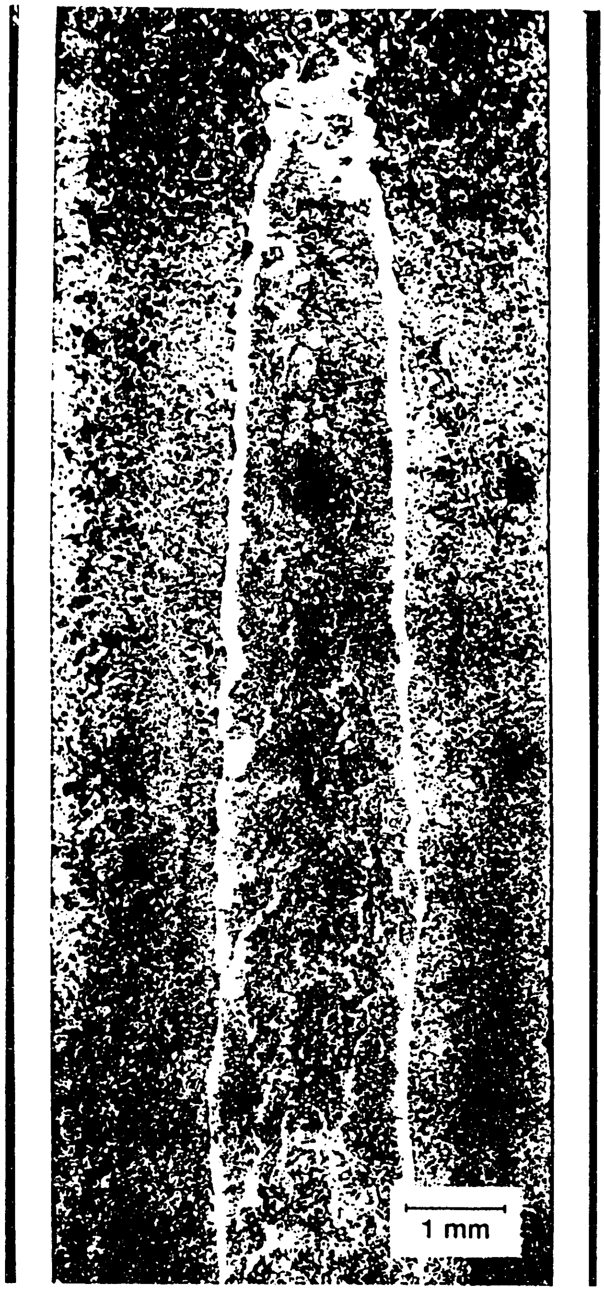
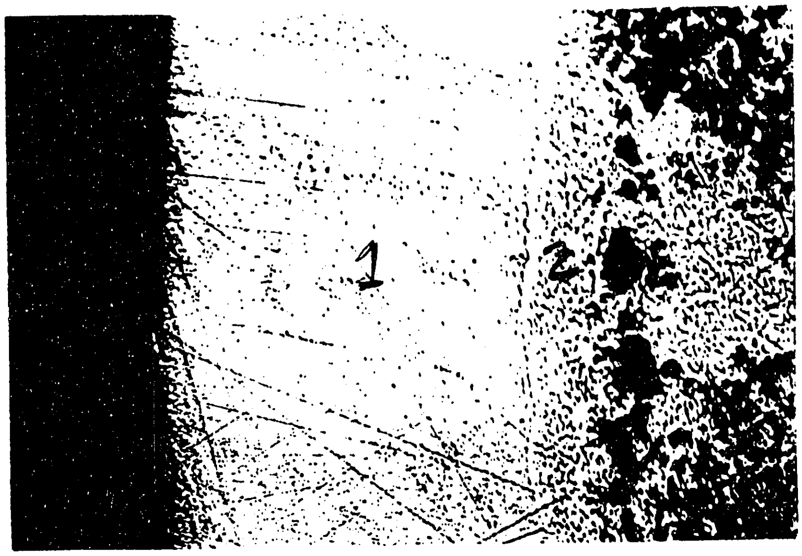
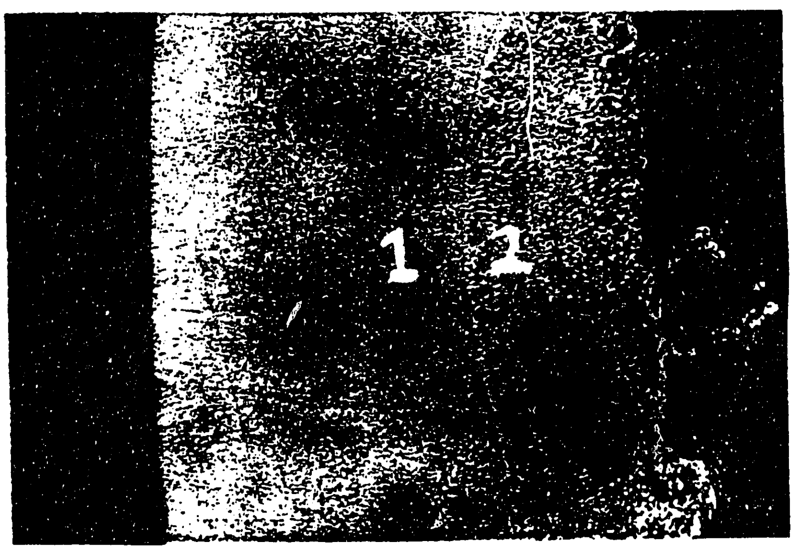


Fig. 7. Longitudinal Section Near the Fuel Top (0.94 ≤ X/L ≤ 0.99) of the FM-5 Fuel Element Showing Low-density Fuel Near the Top, Fuel Restructuring and Alloy Constituent Redistribution, But No Evidence of Fuel Surface Liquefaction.



(a)



(b)

Fig. 8. Fuel/Cladding Interaction-Induced Matrix Dissolution in the HT9 Cladding: (1) Intact Cladding; (2) Reacted Cladding After (a) 1-h FBTA Test at 780°C in a Sibling Fuel and (b) After 112 min. in an FM-2 Fuel-Element Section at the Same Temperature. (150X)

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