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Fuel failure and fission gas release in high burnup PWR fuels under RIA conditions

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Abstract

To study the fuel behavior and to evaluate the fuel enthalpy threshold of fuel rod failure under reactivity initiated accident (RIA) conditions, a series of experiments using pulse irradiation capability of the Nuclear Safety Research Reactor (NSRR) has been performed. During the experiments with 50 MWd/kg U PWR fuel rods (HBO test series; an acronym for high burnup fuels irradiated in Ohi unit 1 reactor), significant cladding failure occurred. The energy deposition level at the instant of the fuel failure in the test is 60 cal/g fuel, and is considerably lower than those expected and pre-evaluated. The result suggests that mechanical interaction between the fuel pellets and the cladding tube with decreased integrity due to hydrogen embrittlement causes fuel failure at the low energy deposition level. After the pulse irradiation, the fuel pellets were found as fragmented debris in the coolant water, and most of these were finely fragmented. This paper describes several key observations in the NSRR experiments, which include cladding failure at the lower enthalpy level, possible post-failure events and large fission gas release. © 1997 Elsevier Science B.V.

1. Introduction

In order to achieve prudent utilization of natural resources and financial advantage with longer refueling cycles, the discharged burnup of commercial power-producing light water reactor (LWR) fuels has been increased in recent years. The burnup limit in Japan has been increased from 39 to 48 MWd/kg U for PWRs and 50 MWd/kg U for BWRs, and further increase of the limit to 55 MWd/kg U is in consideration. As for normal operating conditions, acceptable performance of the fuel has been shown in irradiation programs, operating experiences and analyses. However, extensive investigation for the behavior of the high burnup fuel during off-normal and accident conditions, in particular during reactivity initiated accident (RIA) conditions, is needed. Recent in-pile experiments performed in the Nuclear Safety Research Reactor (NSRR) of the Japan Atomic Energy Research Institute (JAERI) and in the CABRI test reactor of the Institut de Protection et de Sûreté Nucléaire (IPSN) show that the fuel failure may occur at a fuel enthalpy lower than expected.

Current Japanese safety evaluation guideline for reactivity initiated events defines an absolute limit of fuel enthalpy as 963 J/g fuel (230 cal/g fuel) to avoid mechanical forces generation. The guideline also defines an allowable limit of fuel enthalpy for fuel design as 272 to 712 J/g fuel (65 to 170 cal/g fuel), as a function of difference between rod internal and external pressures. When fuel rod internal pressure is lower than external pressure, as is in PWR, the limit is 712 J/g fuel. The guideline was established by the Nuclear Safety Commission of Japan in 1984 based mainly on the results of the NSRR experiments, but all of the NSRR data used were limited to those derived from the experiments with fresh, un-irradiated fuel rods. For this reason, the current guideline adopted the energy deposition at the cladding failure of 356 J/g fuel (85 cal/g fuel) in the SPERT 859 experiment as a provisional failure threshold of pre-irradiated fuel rod; and this failure threshold is used to evaluate the number of failed pre-irradiated fuel rods, and to assess the source term regarding fission gas release in a postulated RIA. For commercial LWR plants to be licensed, the

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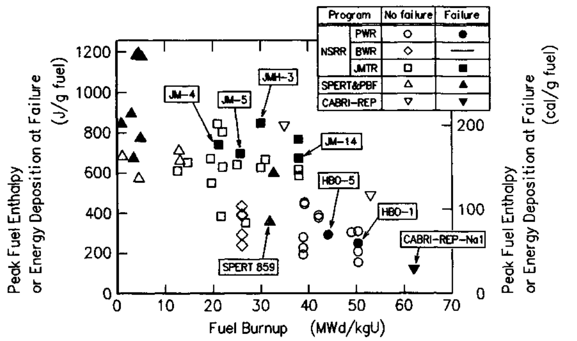

Fig. 1. Peak fuel enthalpy and energy deposition at fuel failure during transients in in-pile RIA experiments as a function of fuel burnup of subjected test fuels. Open symbols denote peak fuel enthalpy in an experiment resulting in no failure, and solid symbols denote energy deposition at failure in an experiment resulting in fuel failure.

safety evaluation must show that the events yield acceptable consequences.

In the HBO-1 test in the NSRR and the CABRI REP-Na 1 experiment an energy deposition appeared at a fuel failure of 250 J/g fuel (60 cal/g fuel) for 50 MWd/kg U PWR fuel and 125 J/g fuel (30 cal/g fuel) for 63 MWd/kg U fuel [1–4]. Fig. 1 summarizes the existing data of peak fuel enthalpy and energy deposition at fuel failure during transients in in-pile RIA experiments as a function of fuel burnup of subjected test fuels. The data points representing the data of the HBO-1 and CABRI REP-Na 1 tests suggest decreased failure threshold in the high burnup region. This paper describes the fuel failure, post-failure events and fission gas release observed in the NSRR experiments. The test fuel rods subjected to the pulse irradiation in the NSRR include segmented fuel rods refabricated from full size fuel rods of commercial power reactors and short fuel rods pre-irradiated in the Japan Materials Testing Reactor (JMTR) of JAERI. Fuel failure as observed in the HBO-1 test was reproduced in the recent HBO-5 test with 44 MWd/kg U PWR fuel. The cladding failure mode in the HBO-5 test is similar to that of the HBO-1 test, and the energy deposition at failure is a little higher in the HBO-5 test. The post-test evaluation and fuel examination for the HBO-5 test are not completed, so it is a little premature to describe the results from the HBO-5 test here.

2. Pulse irradiation in the NSRR

The NSRR is a modified TRIGA-ACPR (annular core pulse reactor) of which salient features are the large pulsing power capability and large (22 cm in diameter) dry irradiation space located in the center of the reactor core which can accommodate a sizable experiment. The shape of the NSRR power history depends on the inserted reactivity, and the smaller pulse becomes broader. While the full width at half maximum in a \$4.6 pulse is 4.4 ms, that in a \$3.0 pulse is 6.9 ms. The capsule used in the pulse irradiation experiment is a double-container system for the irradiated fuel rod test in the NSRR. The inner capsule is a sealed pressure vessel of 72 mm in inner diameter and 680 mm in height. The capsule contains an instrumented test fuel rod with stagnant coolant water at atmospheric pressure and ambient temperature. During a pulse irradiation experiment, cladding surface temperatures at three elevations, coolant water temperature and capsule internal pressure at the bottom of the inner capsule are measured. In some experiments, sensors for axial elongations of pellet stack and cladding tube are also instrumented.

In a series of the irradiated PWR fuel experiments, four different test fuels have been refabricated from full-size commercial reactor fuels, and subjected to the pulse irradiation in the NSRR. Fuel burnup and linear heat generation rate (LHGR) during the base-irradiation (the irradiation in each commercial reactor or the JMTR) are listed in Table 1. Preceding to the extension of PWR fuel burnup limit from 39 to 48 MWd/kg U, the lead use program of high burnup fuel had been performed in the Ohi unit #1 reactor. The HBO test fuel had been irradiated in this program, and the fuel burnup reached 50.4 MWd/kg U. It should be noted that the HBO fuel was not newly designed and manufactured for the high burnup application. The radial distance between the cladding inner surface and fuel pellet (P/C gap) listed in the table is obtained from metallography for arbitrary horizontal cross-section (round slice). As can be seen in this table, the P/C gap of the HBO test fuels is smaller than those of the other test fuels

Table 1
Test fuel rods subjected to the NSRR pulse irradiation experiments

Test fuel ID	Reactor	Initial enrichment (%)	Irradiation cycle	Fuel burnup (MWd/kg U)	LHGR at last cycle (kW/m)	Radial P/C gap (mm)
HBO	Ohi#1	3.2	4	50.4	15.4	< 0.01
OI	Ohi#2	3.2	2	39.2	20.5	0.02
MH	Mihama#2	2.6	4	38.9	19.3	0.02
GK	Genkai#1	3.4	3	42.1	19.8	0.02
JM	JMTR	10	15 to 25	12 to 40	about 25	0.085
JMH	JMTR	20	15 to 25	12 to 40	about 25	0.085

Table 2
Pulse irradiation conditions of irradiated PWR fuel experiments and selected JM and JMH experiments

Test ID	Fuel burnup (MWd/kg U)	Inserted reactivity ($)	Energy deposition (J/g fuel)	Peak fuel enthalpy (J/g fuel)	Remarks
HBO-1	50.4	4.6	390	305	large axial cracking
HBO-2	50.4	3.0	215	157	
HBO-3	50.4	4.6	397	310	
HBO-4	50.4	3.6	279	211	
OI-1	39.2	4.5	571	444	
OI-2	39.2	4.6	581	453	
MH-1	38.9	3.4	262	196	
MH-2	38.9	3.8	301	228	
MH-3	38.9	4.3	363	280	
GK-1	42.1	4.3	505	389	
GK-2	42.1	4.2	490	377	
JM-4	21.2	3.58	986	743	12 small defects
JM-5	25.7	3.37	934	697	23 small defects
JM-14	38	3.59	890	670	large axial cracking
JMH-3	30	3.47	1130	850	large axial cracking

since creep down of the cladding of the HBO test fuels exceeded that of the other test fuels. The information regarding the test fuel pre-irradiated in the JMTR is also listed in Table 1. Because of the limitation of the NSRR pulsing capability and the low residual fissile in the irradiated commercial reactor fuel, the maximum fuel enthalpy in the experiments with the irradiated commercial reactor fuels is restricted to 500 J/g fuel (120 cal/g fuel) or lower. On the other hand, the fuel rods subjected to the pre-irradiation in the JMTR, JM and JMH test fuels, contain the fuel initially enriched to 10% (JM) or 20% (JMH). This relatively high initial enrichment of the JM test fuel realizes the higher fuel enthalpy during the pulse irradiation in the NSRR. The P/C gap of the JM test fuel keeps almost the same value through the pre-irradiation, since the JM test fuel is irradiated in the capsule containing helium gas at atmospheric pressure. The details of the NSRR, the test scheme and the test fuel rods were previously reported in the documents [1,2,5,6]. The pulse-irradiation conditions including the energy deposition and peak fuel enthalpy are listed in Table 2. Because of high burnup and the small number of residual fissile in the HBO fuel, peak fuel enthalpy was restricted to 310 J/g fuel (74 cal/g fuel).

3. Results and discussion

3.1. Cladding failure

A total of forty two experiments, including nineteen commercial LWR fuel tests and twenty JMTR pre-irradiated fuel tests, were performed in the NSRR. Fuel failure occurred in eight of the forty two experiments. Cladding failure modes in these experiments can be categorized in the following manner: (a) vertical cracking over the full length of the fuel active length occurred in high burnup PWR fuel experiments, i.e., the HBO-1 [1,2] and HBO-5 tests; (b) generation of small wall-through defects in the vicinity of the pre-existing local hydride clusters in JM experiments, i.e., JM-4 [5] and JM-5 [6] tests and (c) vertical cracking originating from the local hydride spots observed in JM and JMH experiments, i.e., the JM-14 and JMH-3 tests. Fig. 2 shows the post-test appearance of the fuel rods in the HBO-1, JM-4 and JM-14 tests. The axial crack of the cladding in the HBO-1 test corresponds to the entire region of the fuel stack. The fracture in the experiment is similar to that which occurred by hydride-assisted PCMI (pellet/cladding mechanical interaction) in the SPERT 859 experiment. On the other hand, a number of small defects, twelve in total, were generated in the JM-4 test, and these wall-through defects were distributed over the active region. The long axial crack observed in the JM-14 test propagates through the axially local hydrided spots. Fig. 3 shows horizontal cross-sections in the vicinity of the cracking observed in the HBO-1, JM-4 and JM-14 tests. As for the HBO-1 test, significant hydride deposition below the oxide film in the cladding outer surface and many small cracks vertical to the surface can be seen in the picture. It can be thought that the wall-through crack in the HBO-1 tests originated from one of these crack tips. The crack shows a feature of brittle fracture in the outer region where a number of hydride clusters is deposited. On the other hand, the crack propagates diagonally to a radius in the inner region, and hence shows a typical feature of a ductile fracture. In the JM-4 and JM-14 tests, appearance of the cracks seems similar to that of the HBO-1 test. The cracks show brittle fracture in the outside and ductile in the inside. Although the state of pre-existing hydride deposition is different between the HBO and JM test fuels, as

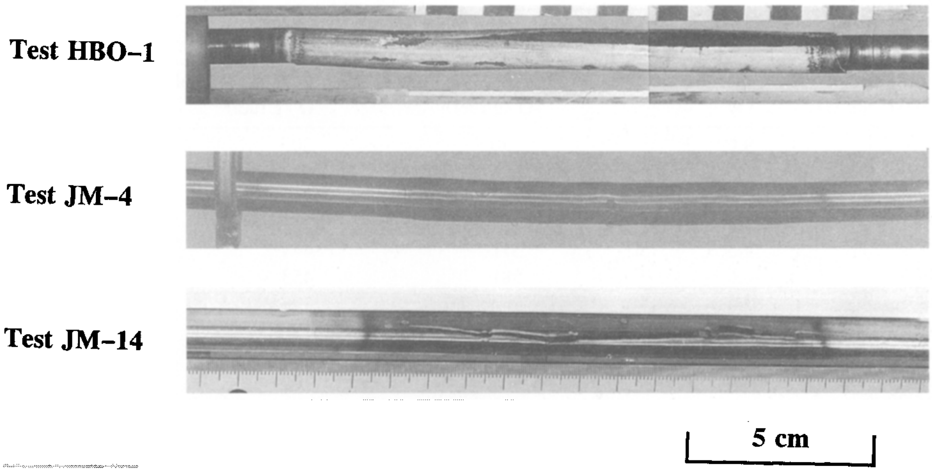

Fig. 2. Post test appearance of the test fuel rods in the tests HBO-1, JM-4 and JM-14. Large axial cracks generated in the tests HBO-1 and JM-14. A number of small wall-through defects appeared in the test JM-4.

shown in Fig. 4, appearance of the cracks suggests strong influence of the pre-existing hydride on the cladding failure in both the HBO and JM experiments. The hydride deposition in the JM test fuels is generated with residual air in the pre-irradiation capsule. In the several experiments, e.g., JM-4 and JM-5 tests, the cracks remain as small wall-through defects since the hydride deposition in the JM test fuel is localized not only radially but also axially and circumferentially. However, in the JM experiment with higher burnup or higher peak fuel enthalpy at pulse, e.g., JM-14 and JMH-3 tests, the crack propagates axially through several hydrided spots, and results in the large opening. Fig. 5 illustrates the interrelations of the elevations where the signals due to local hydride were detected during the eddy current test before the pulse; axial profile of the cladding outer diameter after the pulse; and locations where the wall-through defects were found after the pulse of the JM-4 test. The figure indicates that the occurrence of the cladding failure during the pulse irradiation is strongly influenced by the initially existing cladding hydride, since the location of the cladding defects generated during the pulse irradiation correlates well with the elevation where the signals corresponding to the pre-existing hydride were detected. At the elevation corresponding to the axial center of fuel pellet 'D' in the figure, hydride deposition was detected, and the cladding defect (#8) was

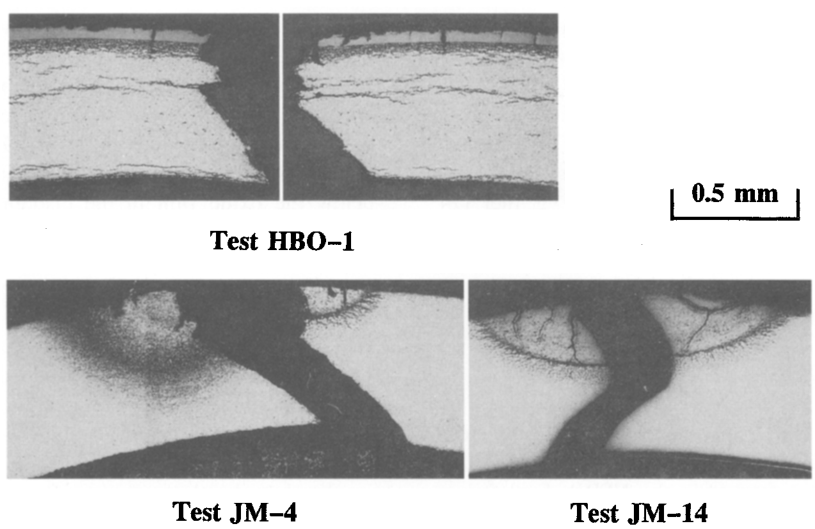

Fig. 3. Horizontal cross-sections in the vicinity of the cracks in the tests HBO-1, JM-4 and JM-14.

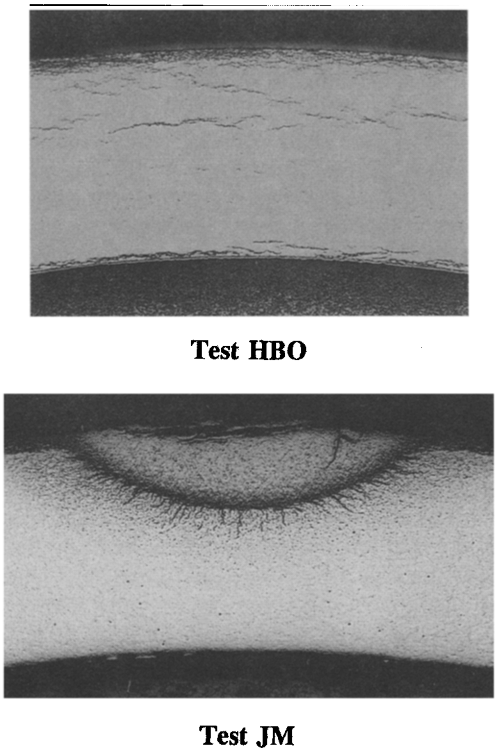

Fig. 4. Pre-existing hydride deposition in the HBO and JM test fuels.

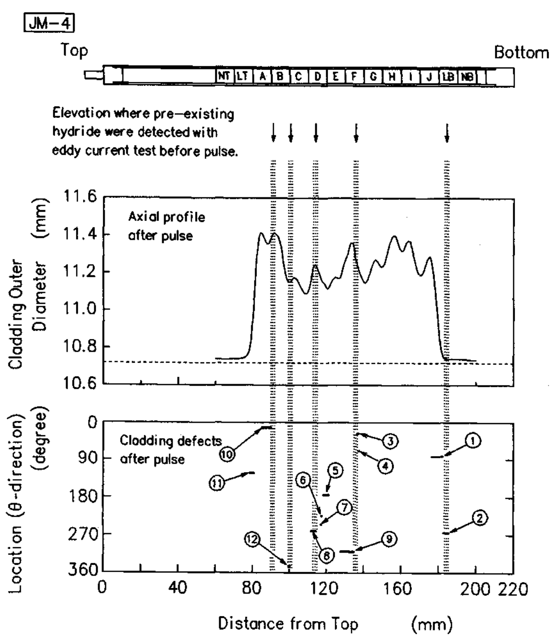

Fig. 5. Interrelations of pre-existing local hydride deposition, post-test cladding outer diameter and cladding defects generated at pulse irradiation in the test JM-4.

generated. At this elevation, the cladding outer diameter is less than 11.3 mm after the pulse. On the other hand, at the elevation of the axial center of pellet 'H' where hydride was not detected, the cladding outer diameter exceeds 11.4 mm, but cladding was not defected during the pulse. These facts indicate that the increase of cladding diameter reaches about 6% due to PCMI with large expansion of the fuel pellets, but the failure does not occur without the existence of the local hydrides.

During the experiments resulting in fuel failure, cladding failure was detected in transient records of capsule internal pressure and/or fuel rod internal pressure as pressure spikes. Transient records of the cladding surface temperature in each experiment show that the temperature remains relatively low at the instant of the cladding failure. As for the HBO-1 test, the cladding surface temperature at failure is about 320 K, and about 370 K in the JM-4 test. The cladding failure of the JM-4 test occurred before departure from nucleate boiling. Occurrence of these cladding failure in the relatively low cladding temperature suggests the failure mode of PCMI in the NSRR experiment.

The cladding failures in the NSRR, CABRI and SPERT programs with irradiated fuels are all believed to be caused by PCMI, assisted by embrittlement of the zircaloy cladding at regions with high local concentrations of hydrides [7]. Quantitative information, however, has not been obtained to define the influence of the hydride precipitation on the embrittlement, and on the cladding failure. Several organizations including JAERI and IPSN have initiated extensive separate-effect tests accordingly, including highly transient burst tests and ring tensile tests with newly developed method. These works will provide correlation between the hydride concentration and distribution, and cladding ductility loss under RIA conditions, including recovery of cladding ductility as a function of time and temperature.

3.2. Post-failure events

Fig. 6 shows transient histories of the capsule internal pressure in the JM-14 test. During the pulse operation, the

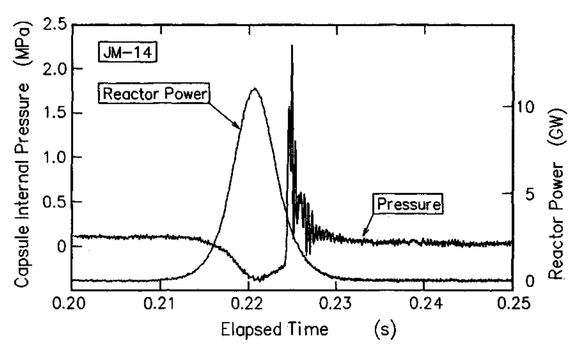

Fig. 6. Transient record of capsule internal pressure during the test JM-14. The pressure spike appeared at cladding failure.

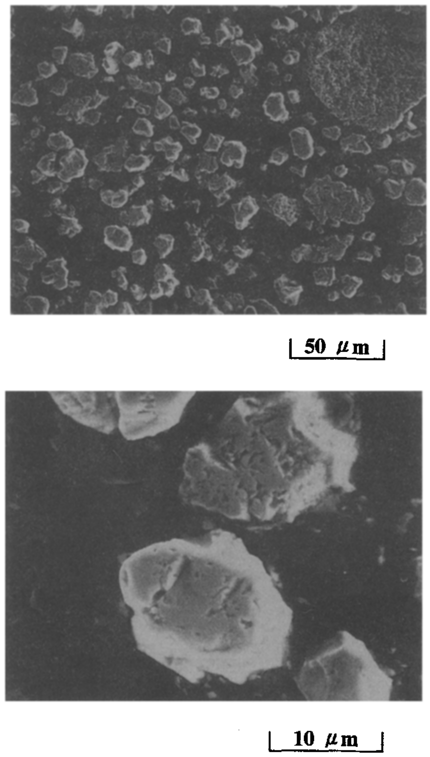

Fig. 7. Cross-sectional view of finely fragmented debris in the test HBO-1.

pressure signal gives a negative value as a noise due to neutron and gamma-ray irradiation. Pressure spikes can be seen in the history, and a peak reaches 2.3 MPa. In the JMH-3 test, the peak pressure of spikes reaches 4.8 MPa at maximum. The pressure spike generation is generally observed in the NSRR experiments resulting in cladding rupture [8]. The cladding failure in the irradiated fuel experiments is generated due to PCMI in the very beginning of the transient when the cladding surface temperature remained low. The pressure spikes caused with cladding rupture and subsequent release of fuel rod plenum gas become large when the cladding temperature remains low at failure.

In the HBO-1 test, all the fuel pellets were dispersed into the capsule water, and were found as fragmented debris [1,2]. Since the collected fuel pellets are finely fragmented, it can be thought that the fuel pellets are expelled from the fractured opening during the pulse. The particle size distribution of the debris shows an occurrence of intensive fragmentation. About 90% of the recovered particles are smaller than 500 μm, and a half or more is smaller than 50 μm. The appearance of the fragmented particles with optical and scanning electron microscopy (SEM) is shown in Fig. 7. During the PIE process, once-molten, spherical particles were not observed.

The predicted fuel temperature of the experiment is about 2600 K at maximum, which is well below the melting point, and hence the fuel pellets of the HBO-1 test have not melted during the experiment. Consequently, the possibility of mechanical energy generation due to violent molten fuel–coolant interaction, or steam explosion, can be neglected. However, fuel fragmentation as observed in the HBO-1 test may become a potential threat concerning fuel coolability, source term, plant contamination, etc. Prompt contact of fuel particles with coolant water may produce high pressure boiling bubbles and cause a pressure surge in PWRs, since the surface area of the finely fragmented fuel particles is considerably large. One of the indications regarding the pressure surge is observed in an NSRR fresh fuel experiment under high pressure and high temperature conditions [9], i.e., test #1206. This experiment was performed with stagnant water coolant at an initial pressure of 7.2 MPa and an initial temperature of 550 K and without gas plenum in the test section. Cladding failed at a relatively low energy deposition, below 800 J/g fuel, and most of the recovered fuel debris was relatively coarse and not once-molten. However, after a sharp pressure spike caused by cladding rupture, a pressure surge, 1.38 MPa increase, was observed in the experiment. In BWRs, such a pressure surge is hardly expected because of pre-existing voids. The pressure surge is not seen in most of the NSRR fresh fuel experiments resulting in vigorous fuel fragmentation, since a free interface exists between the coolant water and capsule plenum gas in the experimental system.

3.3. Fission gas release

Fission gas release to the fuel rod plenum region was destructively measured by rod puncture and gas analysis after the pulse irradiation experiments. The fission gas release during the pulse-irradiation is shown in Fig. 8 as a function of the peak fuel enthalpy. Fission gas release from the HBO fuel during base-irradiation was 0.49%. On the other hand, a significantly large fission gas release oc-

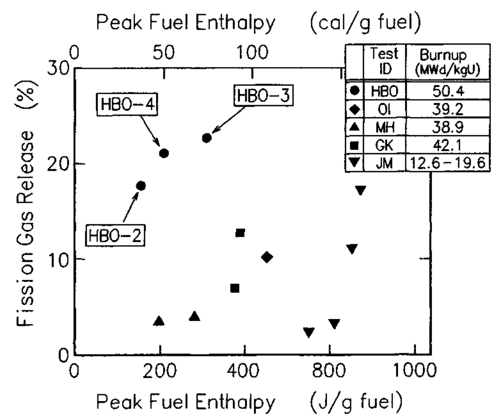

Fig. 8. Fission gas release as a function of peak fuel enthalpy.

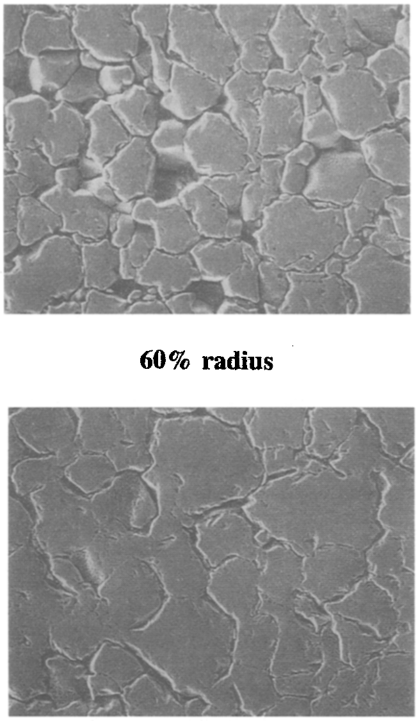

Fig. 9. SEM image of the post-test fuel pellet horizontal cross-section (test JM-4). Grain boundary separation can be seen.

curred in the pulse-irradiation of the HBO-2, -3 and -4 tests [1,2]. The fission gas release is 17.7% even in the HBO-2 test with the peak fuel enthalpy of 157 J/g fuel, and reaches 22.7% in the HBO-3 test. It should be noted that the fission gas release in the HBO-2 test is higher than the release in previous GK-1 experiments with a fuel burnup of 42 MWd/kg U and a peak fuel enthalpy of 389 J/g fuel. The results indicate that the fission gas release during the pulse irradiation depends mostly on the fuel burnup, and the higher fuel burnup correlates with the higher fission gas release.

Grain boundary separation was observed in the JM-4 test, and in the subsequent JM-5 experiment. As can been seen in Fig. 9, a secondary electron image of post-pulse fuel pellets shows the occurrence of significant grain boundary separation in an extensive area. Rapid pressurization of fission gas accumulated in the grain boundaries may cause weakening of the boundaries and subsequent separation, and then result in the expansion of the fuel pellets, fission gas release and fuel fragmentation. The results suggest that, at least, the whole amount of fission gas accumulated in the grain boundary may be released during the pulse irradiation. A preliminary calculation by using FRAPCON-2 code [10] with FASTGRASS module (version of the year 1983) [11] predicts that about 10% of the fission gas accumulates in the inter-granular region of the fuel subjected to the HBO test series. The peripheral 60 μm region of the HBO fuel pellet was characterized as rim region by loss of optically definable grain structure and high concentration of small porosity. The local burnup calculated with RODBURN code [12] reaches 82 MWd/kg U or higher in the rim region where local burnup is enhanced by plutonium production and fissioning. The RODBURN code predicts that about 6% of the fission gas is in the rim region. These preliminary calculations suggest that the total amount of fission gas in inter-granule and in rim regions is less than 16%. The fission gas release measured in the HBO-2, -3 and -4 tests is larger than the predicted value for fission gas in inter-granule and in rim regions. One can hardly expect the release of fission gas, including short-life fission products, from intra-granule during the rapid transient of the NSRR experiment. As stated previously, the calculations currently made are in preliminary stages. The analyses should be upgraded to provide an explanation for the significantly high fission gas release in the HBO experiments.

4. Conclusions

The NSRR experiments suggest possible reduction of failure threshold for high burnup fuels, and indicate that PCMI with expansion of the fuel pellets and decreased cladding integrity lead to the failure. Pre-existing hydride blister in the cladding played important roles in the failure of the rods. Rapid thermal expansion of accumulated fission gas could cause the expansion of fuel pellets and fission gas release, and subsequent fuel fragmentation to extremely small particles. The significantly large fission gas release and the fuel fragmentation producing extremely fine particles indicate that the grain boundary separation occurred almost instantaneously during the transients.

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