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High temperature behavior of irradiated mixed nitride fuel during heating tests

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Abstract

Heating tests for U–Pu mixed nitride fuel irradiated in the experimental fast reactor, "JOYO" were performed for evaluation of fission gas release and nitride decomposition behavior. The gas release onset temperature of irradiated nitride fuel is greater than 1700 °C, which is higher than that of oxide fuel at the same heating rate. A high temperature transient may lead to a permanent expansion of the fuel, giving it a lower density than that of the Pb–Bi coolant. Fission gas release in irradiated nitride fuel does not tend to occur at similar temperatures to the decomposition of the fuel. Thus, in the scenarios of a core damage accident for a Pb–Bi cooled fast reactor, this decomposition behavior must be taken into consideration.

Keywords: Nitride materials; Microstructure; Metallography; Scanning electron microscopy

1. Introduction

In the feasibility study on commercialized Fast Reactor Cycle Systems in Japan, a uranium–plutonium mixed nitride fuel with good thermal properties was selected as a candidate fuel form [1] for which conceptual design studies have been performed. Fast reactor design candidates suitable for nitride fuel include middle and small size reactor cores using Pb–Bi alloy coolant and pellet type fuel. In safety studies of these concept reactors, it is quite important to recognize the behavior of the nitride fuel under abnormal fuel temperatures such as in core damage accidents (CDAs). This is because a large amount of fission gases could remain in the nitride fuel due to the lower irradiation temperature experienced as compared to that for oxide fuel, and these gases might be rapidly released as the temperature rises during CDAs. Moreover, if the temperature exceeds 2000 °C, decomposition of the nitride fuel may occur [2]. In some high temperature accidents the cladding will melt, releasing fuel into the coolant. The fuel may sink or float according to whether it is more or less dense than the coolant. However, almost no fission gas release data from irradiated mixed nitride fuel under abnormal temperature conditions have been obtained. The same holds true for data concerning decomposition behavior.

JAEA has studied the applicability of nitride fuel for fast reactors by heating tests using uranium–plutonium mixed nitride fuels irradiated in the experimental fast reactor, "JOYO." In this work, the fission gas release and the decomposition behaviors were evaluated for the safety study of a Pb–Bi cooled fast reactor. The gas release behavior and metallographic observations during and after the tests are shown and discussed.

2. Experimental procedure

The characteristics and irradiation conditions of nitride fuel pellets are shown in Table 1. Results for other post irradiation examinations (PIEs) for these fuel pellets have been given in the literature [3]. A pellet was removed from the fuel pin and the cladding was cut away to obtain test specimens (see Fig. 1). The test specimens were heated in an induction heating furnace in an atmosphere of flowing high-purity Ar carrier gas at 0.1 MPa. Details of the heating apparatus can be seen in the literature [4]. The heating tests were designated Nifti-1 and Nifti-2 (Nifti: nitride fuel heating test after irradiation), using fuel samples of approximately 5 and 3 g, respectively. In Nifti-1, the temperature was slowly elevated to the maximum of 2400 °C to allow a detailed observation of the fission gas release and decomposition behavior (the heating rate is 20 K/min above 1000 °C). In Nifti-2, a relatively rapid heating rate (approximately 20 K/s) was applied from 1700 °C (equivalent to the center line temperature under irradiation) to 2500 °C, which simulated the transient timescale, "slow transient overpower (TOP)." During the rise to the start temperature 1700 °C in the Nifti-2 test, observations of fission gas release behavior were attempted during holds at constant temperatures of 1500, 1600 and 1700 °C.

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Fig. 1. The irradiated mixed nitride fuel after removing cladding.

During the tests, the released gases were analyzed "in situ" in the test rig by gas mass spectrometry, and then the concentrations of released gases were evaluated as a function of temperature. At the same time, a portion of the released gas was intermittently analyzed utilizing gas chromatography to calibrate the in situ gas concentration data. Details of analysis systems have been given in the literature [4]. After these heating tests, metallographic observations and EPMA for these specimens were carried out to get microstructures and elemental distributions.

3. Results and discussion

3.1. Fission gas release behavior

In Nifti-1, concentration profiles of fission gases and nitrogen in Ar carrier gas were obtained as shown in Fig. 2. The fission gas release begins at a heating temperature of approximately 1730 °C. During Nifti-2, the profiles were obtained as shown in Fig. 3. In this case, only a small amount of fission gas was released at temperatures below 1700 °C. Fission gas release behavior depends on the irradiation condition and heating pattern. The release onset temperature of irradiated oxide fuels is approximately 1400 °C [5] at a heating rate similar to that used in Nifti-2. The higher release temperatures for nitride indicate that nitrides may retain fission gases better than oxides do. In particular, it was reported from pin puncture test results that this irradiated nitride fuel pin had a low fission gas release rate during irradiation (approximately 3.3% for the total of fission

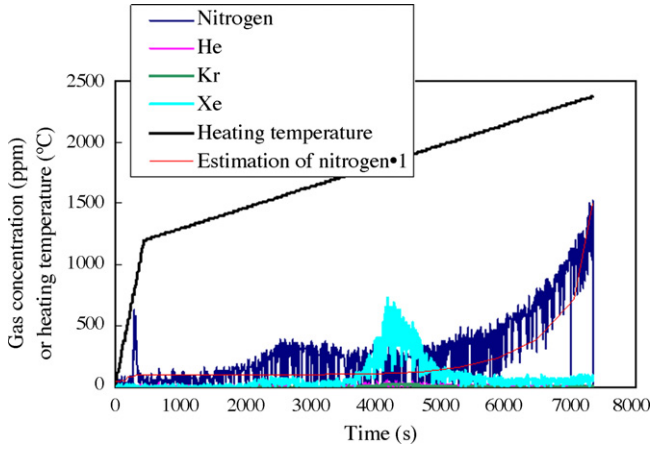

Fig. 2. Heating temperature and released gas concentration profiles during Nifti-1 (maximum temperature = approximately 2400 °C, (*1) these data are relative value from calculated data).

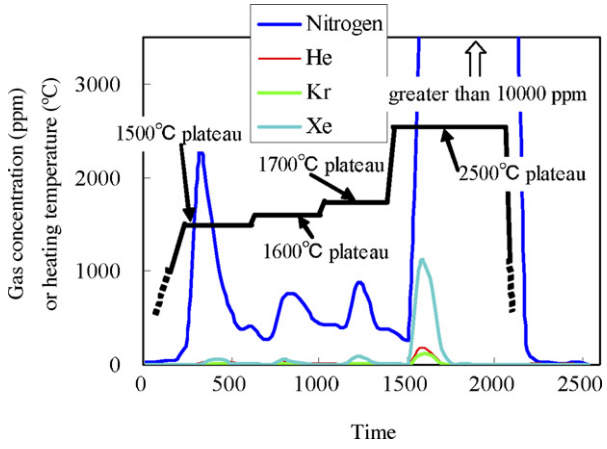

Fig. 3. Heating temperature and released gas concentration profiles during Nifti-2. The gas concentration profile is late for the temperature profile by approximately 100 s (maximum temperature = approximately 2500 °C).

gases [3]). If the onset temperature correlates with fuel fracture toughness and the ease of fission gas bubble growth, this difference between nitride fuels and oxide fuels may result from the difference of the mechanical properties. The fracture surface

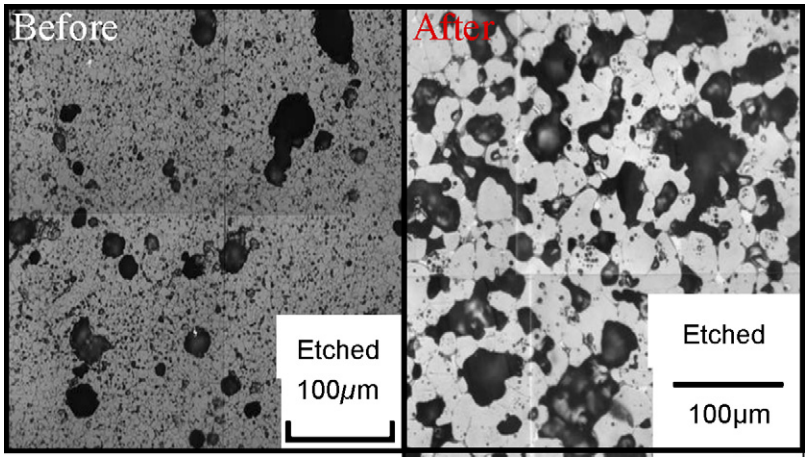

Fig. 4. Metallographic images obtained before and after Nifti-1 (these samples were taken in almost the same radial position ($r/R = 0.5$)).

Table 1
Fuel characteristics and irradiation conditions

Fuel characteristics	
N/*M*	1.00
Diameter	7.28 mm
Pu/(Pu + U)	18.6 wt%
U235 enrichment	19.6 wt%
Density	84.8%T .D.
Stack length	200 mm[a]
Irradiation conditions	
Burn up	Approximately 4 at%
Max fuel	Approximately 1700 °C (pellet center)
temperature	Approximately 1400 °C (pellet surface)

[a] A normal stack length for the "JOYO" MK-II is 550 mm. The fuel core of this fuel pin is shortened.

energy and fracture toughness [6] of nitride fuels are approximately 2.0 J/m^2 and 1.0 MN/m$^{3/2}$, respectively, similar to those of oxide one [7]. The Young's modulus of nitride fuels is more than 250 GPa [8], compared to approximately 200 GPa for oxide fuels [7]. Hence, the high fission gas release onset temperature of the nitride fuel could be caused by slow growth of fission gas bubbles in the deformation-resistant fuel. Alternatively, the difference may be due to slower migration of gas atoms and/or gas bubbles in the nitride fuel.

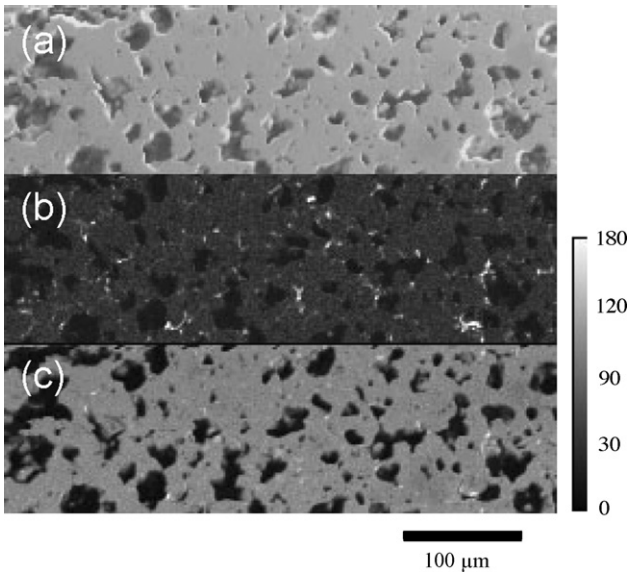

Fig. 5. EPMA of the Nifti-1 specimen: (a) secondary electron image, (b) Rh and (c) Pu.

Fig. 4 shows the metallographic images of a specimen before and after Nifti-1. The quite small pores existing prior to the heating test were able to connect with each other and grow through heating to become larger. Therefore, the fuel density after heat-

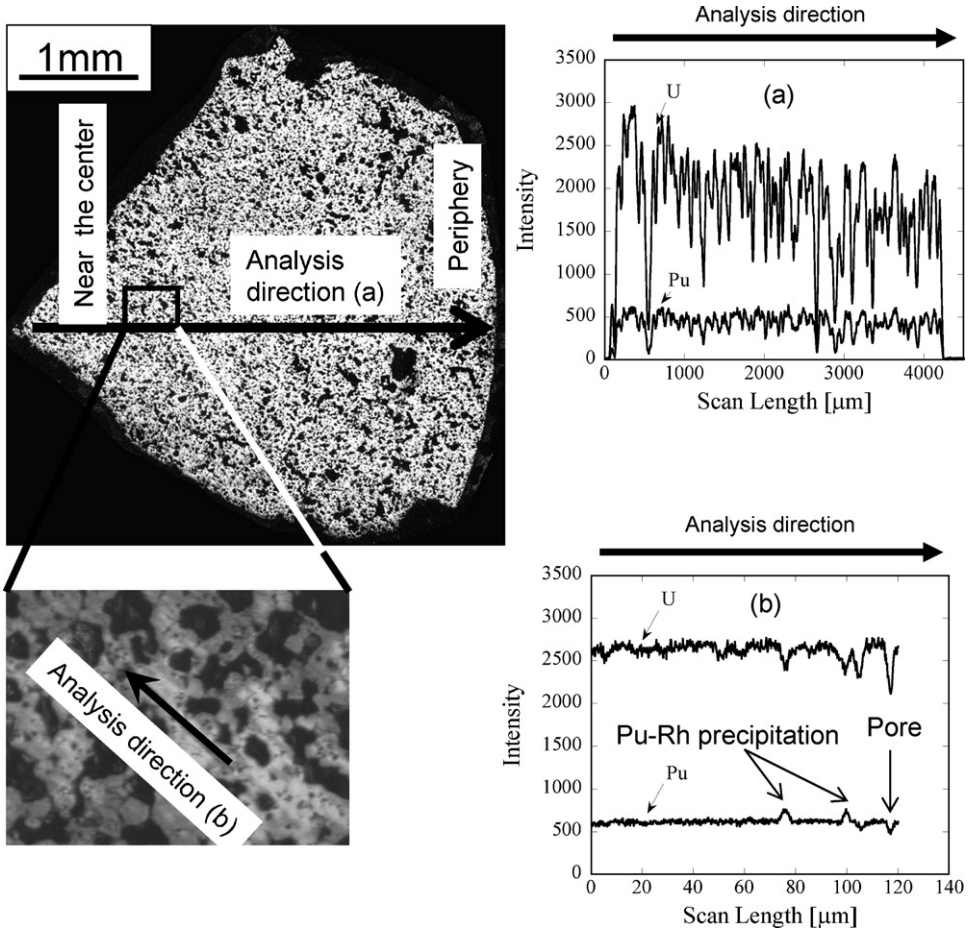

Fig. 6. EPMA of the Nifti-1 specimen for (a) macroscopic and (b) microscopic scale.

ing decreased to less than 9 g/cm^3, smaller than the coolant density (10 g/cm^3). This pore coalesce caused from behavior as mentioned below. In the irradiated fuel, fission gas was pressurized in the bubble, especially in "deformation-resistant fuel" like nitride fuel. The high temperature and no cladding material during the heating test made the fuel soften without pressure constraint. Therefore, the bubbles are able to expand, which might become the lower density of fuel. The fuel pellets could float above the coolant during CDA with cladding melting. Thus, in the scenarios of a core damage accident for Pb–Bi cooled fast reactor, this behavior must be taken into consideration. On the other hand, in the Nifti-2 specimen, there are still small pores after heating. This implies that the pore size and number are dependent upon the heating rate and time. In other words, in Nifti-2, the heating time was not enough for pore growth, although the temperature was higher than in Nifti-1.

3.2. Nitride decomposition and metal inclusion behaviors

Concerning the relationship between the fission gas release and nitrogen release by decomposition, in Nifti-1, the main nitrogen release begins at approximately 1900 °C, but does not correlate with the fission gas release as seen in Fig. 2. The equilibrium nitrogen partial pressure over uranium nitride shown in ref. [9] will be approximately 5×10^{-6}–10^{-8} MPa (50–0.01 ppm under a total pressure of 0.1 MPa). The impure nitrogen concentration of 1 ppm for the Ar carrier gas falls within this range. Therefore, the decomposition behavior of nitride fuel proceeded in accordance with the thermochemical equilibrium process. Here, for comparison, the calculated partial pressure (relative value) of nitrogen will be presented in Fig. 2 also [10], where the increasing aspect is similar to that of measured profile.

EPMA for the Nifti-1 specimen shows some inclusions containing Pu and Rh as seen in Fig. 5. We have confirmed that U concentration is very low at such inclusions (see Fig. 6), nevertheless, precipitation of Rh was observed with U prior to the heating test [11]. In addition, neither Pd nor Ru was detected. The concentration profiles of U and Pu along the fuel radius are shown in Fig. 6 for macroscopic and microscopic scales. As seen in Fig. 6(a), the intensity of U near the periphery is lower than that near the center, unlike that of Pu. The Pu–Rh precipitation is observed in Fig. 6(b) as well as in Fig. 5. From these data, it is possible to conclude that the nitride fuel starts to decompose near the periphery as has been reported in other studies [12]. U would then react with oxygen impurities (approximately 1 ppm) in the carrier gas to form a gaseous chemical species such as UO_3 more easily than Pu with the accordance of thermochemical consideration [10]. Pu could remain in the fuel matrix or form precipitates with Rh. However, it is remarkable that there is no evidence of an escape of U at other "free surfaces" such as other edges of "sector form" in Fig. 6. We might speculate that this shape is formed after heating by fracture during fuel treatment.

4. Summary

The heating tests for U–Pu mixed nitride fuel irradiated in the experimental fast reactor, "JOYO" were performed. Gas analyses during the tests, metallography and EPMA for heated specimens after the tests produced the following results and discussion.

- The fission gas release onset temperature of irradiated nitride fuel is higher than that of oxide fuel at same heating rate. This difference may result from the different mechanical properties of these fuel types.
- The fuel density may become lower than that of the Pb–Bi coolant by the connection and growth of fission gas bubbles in irradiated fuel in some CDAs with melting cladding materials and without restraint of the cladding.
- Fission gas release in irradiated nitride fuel does not tend to occur at similar temperatures to the decomposition of the fuel. The onset of nitrogen release is near when the nitrogen partial pressure elevates in a thermochemical estimation, which indicates that the decomposition behavior proceeds in accordance with thermochemistry.
- U may leave the fuel more easily than Pu, having reacted with traces of oxygen in the carrier gas and become a gaseous chemical species such as UO_3.

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