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Effects of pellet microstructure on irradiation behavior of UO_2 fuel

R. Yuda [a], H. Harada [b], M. Hirai [a], T. Hosokawa [a], K. Une [a,*], S. Kashibe [a], S. Shimizu [c], T. Kubo [d]

[a] *Nippon Nuclear Fuel Development Co. Ltd., Higashi, Ibaraki-gun, 2163 Narita-cho, Oarai-machi, Ibaraki-ken 311-13, Japan*
[b] *Tokyo Electric Power Co., 4-1 Egasaki-cho, Tsurumi-ku, Yokohama 230, Japan*
[c] *Toshiba Corp., 8 Shinsugita-cho, Isogo-ku, Yokohama 235, Japan*
[d] *Hitachi Ltd., 1-1 Saiwai-cho, 3-chome, Hitachi-shi, Ibaraki-ken 317, Japan*

Abstract

In-reactor tests and post-irradiation examinations (PIEs) were performed for standard and large-grained pellets with and without additives being soluble in a matrix and/or precipitated in a grain boundary, to confirm the effects of large grain structure on decreasing fission gas release (FGR) and swelling and to evaluate the influence of the additives in the matrix/grain boundary on them. The standard and large-grained pellets were loaded into small-diameter rods equipped with a pressure gauge. These rods were irradiated to about 60 GWd/t U at a linear heat rate of about 30–40 kW/m in the Halden reactor and then subjected to PIEs. Large-grained pellets showed a smaller FGR compared with standard pellets. Post-irradiation annealing tests suggested that swelling during transient power was decreased for large-grained pellets, except for those with additive enhancing cation diffusion. © 1997 Elsevier Science B.V.

1. Introduction

The increase in fuel rod internal pressure caused by fission gas release (FGR) and the increase in pellet-cladding interaction (PCI) by swelling are key problems affecting fuel reliability at high burnup. A large-grained pellet has been considered to suppress FGR and swelling, since, in principle, both are rate-controlled by gas atom diffusion from grain interiors to grain boundaries.

The large-grained pellet can be produced by controlling powder characteristics [1] and/or sintering conditions [2–5], and by using additives [6,7]. Some additives are soluble in the matrix and enhance cation diffusion, other additives form a liquid phase during sintering and deposit in the grain boundary, changing its properties. Many studies have elucidated the effects of grain size and additives on fission gas release and bubble swelling. According to a post-irradiation annealing experiment using 23 GWd/t U fuels in a previous study, FGR and bubble swelling for large-grained fuel were distinctly lower than those for standard fuel [8]. On the other hand, an additive such as TiO_2 and Nb_2O_5 accelerated FGR and swelling by enhancing fission gas diffusivity [8,9]. However no systematic assessment was made on the grain size effect for non-additive and additive-doped fuels irradiated to high burnup.

In the present study, FGR and swelling during base irradiation and transient power for large-grained fuel with and without additives, which were irradiated to about 60 GWd/t U at a linear heat rate of about 30–40 kW/m, were examined by PIEs incorporating the same annealing technique as previous studies [8,10].

2. Experimental

2.1. Fuel and rod design

The main fuel characteristics are shown in Table 1. In this study, standard fuel and four types of large-grained fuels were prepared: (1) standard fuel, (2) non-additive large-grained fuel, (3) titania-doped large-grained fuel, (4) alumino-silicate-doped large-grained fuel, (5) titania-silicate-doped large-grained fuel.

* Corresponding author. Tel.: +81-29 266 2131; fax: +81-29 266 2589; e-mail: une@nfd.co.jp.

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The titania-doped and titania-silicate-doped fuels had enhanced cation diffusivity in the matrix compared with standard fuel. The alumino-silicate-doped and titania-silicate-doped fuels differed from standard fuel regarding the grain boundary properties. All fuels had 13% enrichment and a small diameter of 5.5 mm, which allows rapid burnup accumulation and a similar fuel temperature to that of fuels with normal dimensions.

The zircaloy cladding tubes had a small diameter (O.D. = 6.53 mm, I.D. = 5.61) and a Zr-liner with thickness of 50 μm.

Each rod had a fuel active length of about 230 mm and was filled with He of 1 MPa, including two end pellets. The diametral gap width was 110 μm.

2.2. Irradiation

The fuel rods were irradiated to about 60 GWd/t U at the Halden reactor. The fuels were irradiated under a small axial power distribution (peak/average < 1.01). Fig. 1 shows the power history of the standard fuel rod as a typical example. At full reactor power, the average linear heat rate of the rods was kept at about 30–40 kW/m.

2.3. Post-irradiation examinations

The retrieved fuel rods were subjected to post-irradiation examinations (PIEs) after cooling for a half year. The PIEs included:

- Dissolution tests for evaluation of FGR by measuring the retention of ^{85}Kr gas.
- Ceramography, scanning electron microscopic (SEM) observation.
- Electron probe micro-analyses (EPMAs) for evaluation of radial burnup and retained fission gas distributions and confirmation of bonding at the pellet/cladding interface.
- Post-irradiation annealing test for evaluation of fractional burst release and bubble swelling during transient power.

Table 1
Characteristics of pellets

	No.				
	1	2	3	4	5
Material [a]	UO_2	UO_2	UO_2–A1	UO_2–A2	UO_2–A3
U-235 enrichment (%)	13				
Density (%TD)	97.17	96.90	97.79	99.54	99.45
Grain size (μm)	9	51	46	68	135
Diameter (mm)	5.504	5.503	5.501	5.499	5.498
Length (mm)	5.66	5.59	5.53	5.53	5.56
Geometry	flat-ended				

[a] A1: 0.25 wt% alumino-silicate. A2: 0.2 wt% titania. A3: 0.25 wt% titania-silicate.

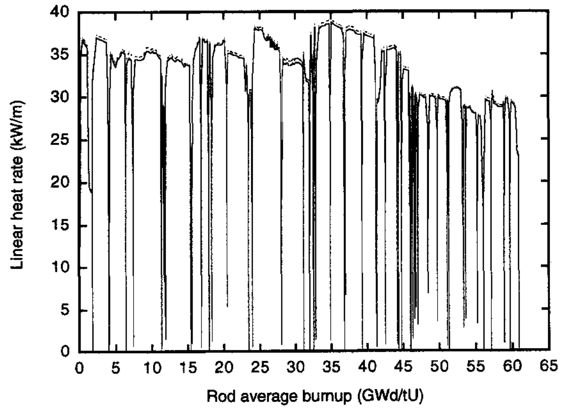

Fig. 1. Power history of the standard fuel rod. ---: maximum, ————: average.

In this test, specimens were heated to 1800°C for 5 h in a reducing atmosphere of dry He–2%H_2. The heating rate at the temperature ramp was 1.7°C/s.

Through these PIEs, we evaluated the effects of a large grain structure on decreasing FGR and swelling and the influence of the additives in the matrix/grain boundary on them.

3. Results and discussion

3.1. Irradiated fuel characteristics

Fig. 2 shows ceramographs of as-etched standard fuel. Crack patterns can be seen in the fuel, which were considered to be formed in power down. Between the pellet and cladding, the bonding layer is observed instead of the gap. U and Zr interdiffusion at the interface was confirmed by EPMA. Almost the same bonding layer is observed in the other fuels. The reason why this bonding occurred is due to severe PCI caused by the bubble swelling and the narrow gap for these fuels.

In the central region (Fig. 2), large intergranular bubbles and metallic inclusions were found, which suggests a high fuel temperature. A similar structure is found in other fuels. Although the grain size increases to about 3 times the initial value in the central region for standard fuel, grain growth does not appear in large-grained fuels.

There are a lot of intragranular bubbles in the periphery and outer part of the middle region as seen in Fig. 2. These bubbles could not be observed in as fabricated fuels. In the other fuels, there are similar bubbles. SEM and TEM observations on the fracture and polished surfaces confirmed the presence of sub-divided grain structure in these areas as shown in our other paper [11]. Usually, these sub-divided grain structures have been seen in the periphery of commercial fuels irradiated to high burnup [12–15]. It is a noticeable point that the sub-divided grain structure is also seen in the middle region. The mechanisms of the sub-divided grain structure formation at the middle region is discussed in our other paper [11].

3.2. Fission gas release

Fig. 3 compares gas pressure history between standard and non-additive large-grained fuel rods. The gas pressures of both rods increased gradually after about 20 GWd/t U and increased stepwise at power down after 30 GWd/t U, which were considered to be caused by PCI [16]. Indeed, the increase in diameter of about 40 μm (0.6%) and ridging, which suggest severe PCI during irradiation, were detected by diameter profile measurements for the standard fuel rod. The non-additive large-grained fuel rod showed a smaller gas pressure, that is FGR, compared with the standard fuel rod before 50 GWd/t U. However the standard fuel rod showed no increase in gas pressure after 50 GWd/t U, which was attributable to blocking up by gap closure caused by severe PCI as described above. In other rods, the gas pressure increased in a similar way to non-additive large-grained fuel rod.

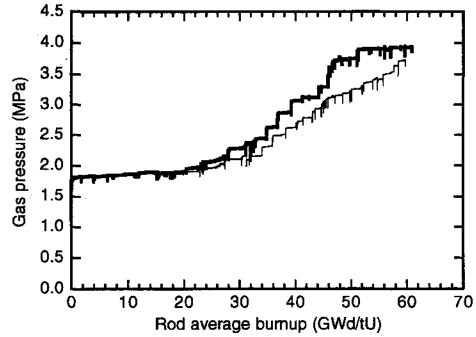

Fig. 3. Comparison of gas pressure history between standard fuel and non-additive large-grained fuel rods. ————: standard, ————: non-additive large-grained.

The FGR was evaluated from a dissolution test instead of a puncture test because of plugging in the active stack region for all fuel rods. Fig. 4 shows FGR as a function of reciprocal grain size. FGRs of the large-grained fuels was

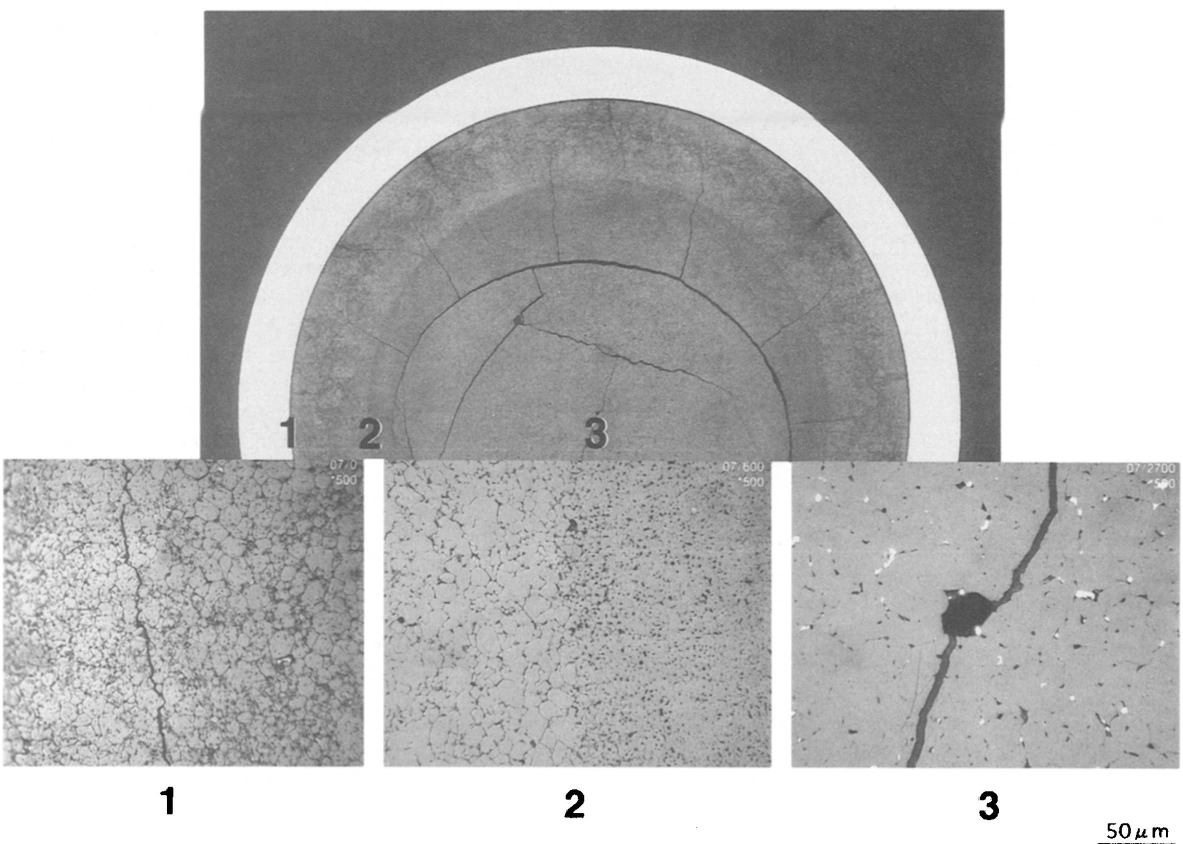

Fig. 2. Ceramographs of standard fuel as etched.

about 70% of the standard fuel value. Under irradiation, FGR, expressed as release to birth ratio (R/B), can be related to the effective diffusion coefficient D by the following approximation of Booth's model [17]:

$$(F/B) \propto D^{1/2}/a, \tag{1}$$

where a is Booth's equivalent sphere diameter. In this study, a is considered to equal roughly the as-fabricated grain size. The grain size effect on FGR was smaller than that predicted by Eq. (1) for all large-grained fuels. In the central region, fission gas is almost completely released regardless of grain size because of the high temperature. On the other hand, fission gas is not released without reference to grain size in the outer region because of the low temperature. In this case, the region in which FGR is affected by grain size is from r/r_0 = about 0.4 to 0.7, which was evaluated from retained Xe profiles. Considering this correction, FGRs of non-additive and alumino-silicate-doped fuels are predicted to be about 50% of that of standard fuel, approaching measured values. Other reasons were considered as follows. Microstructural changes such as grain growth of standard fuel, which decreases the grain size difference from other fuels and such as sub-grain boundary formation, which acts as a short diffusion path for the gases [5], may decrease the advantage of large grain size. For all fuels, the sub-divided grain structure was observed even in the middle region from which fission gas would be released. In addition, the enhancing fission gas diffusivity by the additive [9] may diminish the large grain effect for titania-doped and titania-silicate-doped fuels.

The fractional burst release was evaluated by the same annealing technique as previous studies [8,10]. Fig. 5 shows the fractional burst release as a function of reciprocal grain size.The fractional burst release is considered as FGR from grains to grain boundaries during irradiation and can be also related to grain size by Eq. (1).The non-additive large-grained fuel has half the fractional burst release as standard fuel does. The grain size effect on the fractional burst release was smaller than that predicted by Eq. (1) for the non-additive fuels. This would be attributable to FGR from grain boundaries to outside and microstructural changes such as grain growth for the standard fuel during irradiation. The alumino-silicate-doped fuel shows a little larger fractional burst release than non-additive large-grained fuel. This would be attributable to the increase in the effective surface to volume ratio by melting of the additive on the grain boundaries. The titania-doped fuel shows no grain size effect. For titania-doped fuel, the fractional burst release would be due to a large amount of fission gas, which had accumulated on the grain boundaries due to the enhanced fission gas diffusivity. The titania-silicate-doped fuel also shows about the same tendency as the titania-doped fuel. The fractional burst release of the titania-silicate-doped fuel is about $\frac{1}{2}$ of the value for titania-doped fuel, which roughly corresponds to the reciprocal ratio of grain size of both fuels.

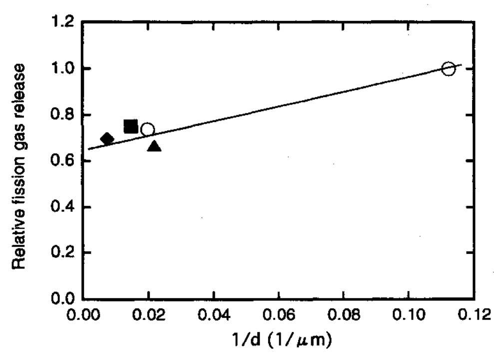

Fig. 4. FGR as a function of reciprocal grain size. ○: non-additive, ■: titania-doped, ▲: alumino-silicate-doped, ◆: titania-silicate-doped.

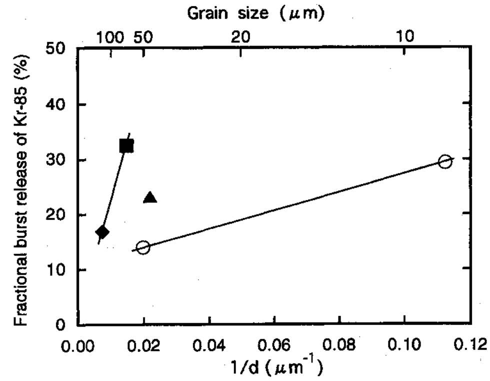

Fig. 5. The fractional burst release as a function of reciprocal grain size. ○: non-additive, ■: titania-doped, ▲: alumino-silicate-doped, ◆: titania-silicate-doped.

From the present results, we confirmed that the large grain structure decreases FGR and the fractional burst release for high burnup fuels irradiated to about 60 GWd/t U. We also confirmed that the additives enhancing cation diffusion accelerate the fractional burst release even in the high burnup fuels.

3.3. Swelling

Scanning electron micrographs after annealing at 1800°C for 5 h are shown in Fig. 6. In all fuels, there are many bubbles which were not observed before annealing. In standard fuel, there are too many bubbles to distinguish individual ones on grain boundaries or in grains. In non-additive and alumino-silicate-doped large-grained fuels, there are large intergranular bubbles and small intragranular bubbles. In large-grained fuels doped with titania or titania-silicate, there are unusually large intragranular bubbles and intergranular bubbles which form much larger grain boundary tunnels of several microns. Bubble swelling after annealing at 1800°C for 5 h was evaluated by the difference in porosity based on ceramographs taken before and after annealing. The swelling data are plotted as a function of reciprocal grain size (Fig. 7). If bubble swelling was mainly caused by saturated intergranular bubbles, the swelling would be simply proportional to reciprocal grain size [18]. Consequently, all fuels swelled not only by intergranular bubbles, but also intragranular bubbles as seen in Fig. 6. However the swelling of non-additive large-grained fuels is about $\frac{1}{4}$ of the value for standard fuel, which roughly corresponds to the reciprocal ratio of grain size of both fuels. This suggests that there is no notable difference between the contribution of intragranular bubbles swelling of both fuels. The swelling of alumino-silicate-doped large-grained fuel is a little more than the value for non-additive large-grained fuel. For alumino-silicate-doped fuels, the melting and vaporization of the additive may contribute to the swelling on the grain boundaries. On the other hand, large-grained fuels with

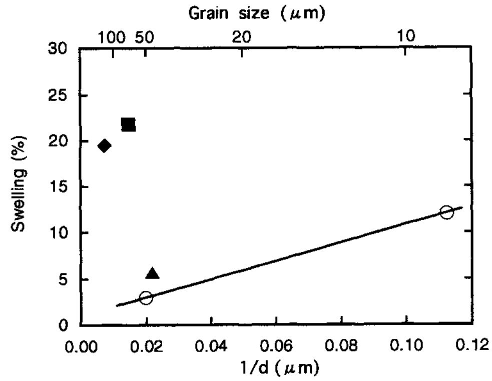

Fig. 7. Swelling as a function of reciprocal grain size. ○: non-additive, ■: titania-doped, ▲: alumino-silicate-doped, ◆: titania-silicate-doped.

doped titania and titania-silicate show much larger swelling compared with non-additive large-grained fuel, which coincides with the results obtained by our previous work [8]

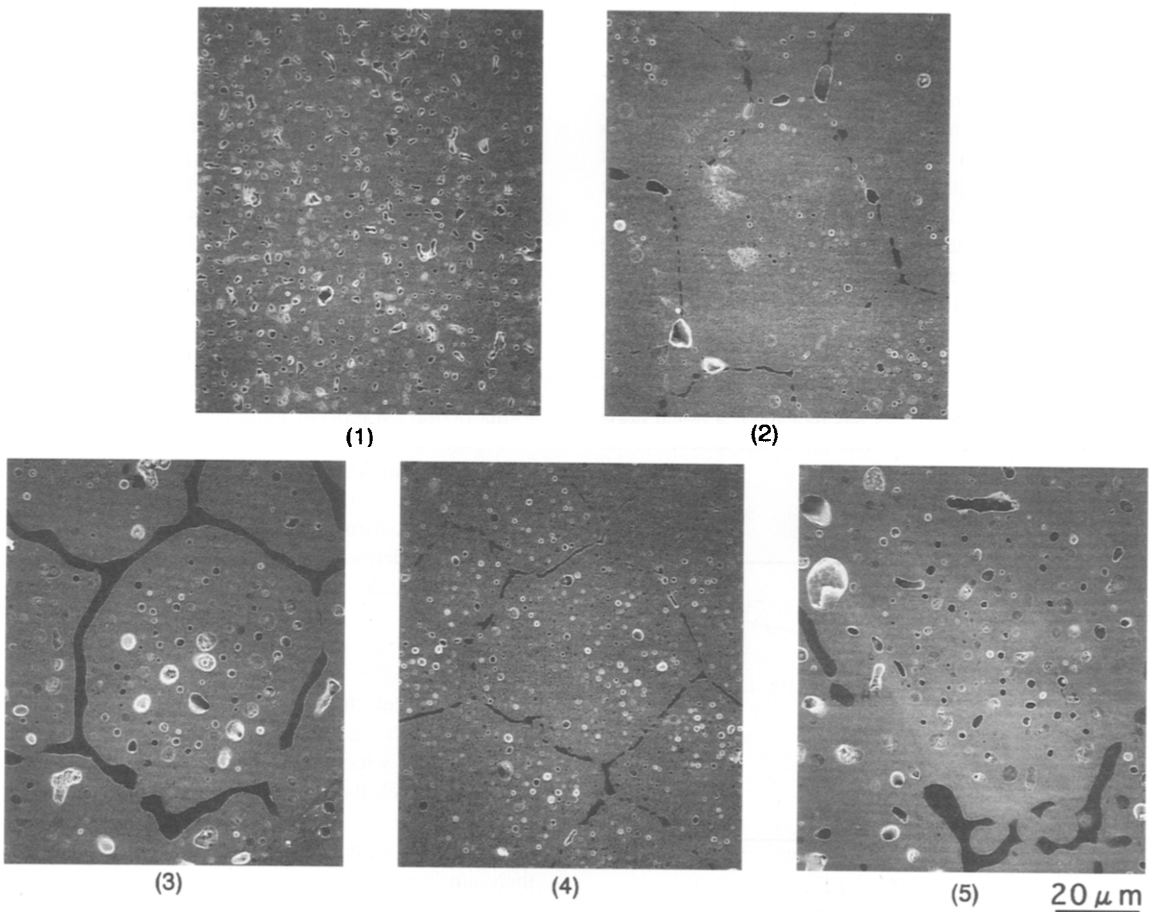

Fig. 6. Scanning electron micrographs of polished surfaces of fuels annealed at 1800°C for 5 h. (1) Standard, (2) non-additive large-grained, (3) titania-doped, (4) alumino-silicate-doped, (5) titania-silicate-doped.

for fuels irradiated to 23 GWd/t U and a simulated bubble swelling technique [18]. The reason is due to the precipitation of coarsened inter- and intragranular bubbles as seen in Fig. 6. The enhanced fission gas diffusivity due to the additive [9] also contributes to the growth of inter- and intragranular bubbles.

These results suggest that the large grain structure decreases swelling during transient power and the additives enhancing cation diffusion, such as titania and titania-silicate, increase the swelling. These results correspond with the above results on the fractional burst release.

4. Conclusions

Standard and large grain pellets with and without additives, which were irradiated to about 60 GWd/t U in the Halden reactor, were subjected to post-irradiation examinations to investigate fission gas release and swelling. The conclusions obtained in the present study were as follows.

(1) The large grain structure decreased FGR during base irradiation, fractional burst release and swelling during transient power for high burnup fuels irradiated to about 60 GWd/t U.

(2) Additives enhancing cation diffusion such as titania and titania-silicate increased the fractional burst release and swelling during temperature ramp even for high burnup fuels irradiated to about 60 GWd/t U.

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