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Journal of Nuclear Materials 352 (2006) 365–371
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Post-irradiation examination on particle dispersed rock-like oxide fuel

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Abstract

To evaluate the irradiation behavior of the rock-like oxide fuel, irradiation experiments were carried out. Three fuels were prepared; a single phase fuel of yttria-stabilized zirconia containing UO_2 (U-YSZ) and two particle-dispersed fuels of U-YSZ particles in spinel or corundum matrix. U-YSZ particle sizes were about 100–200 μm. These fuels were irradiated in Japan Research Reactor No. 3 for about 280 days. Post-irradiation examinations such as visual inspection and γ scanning were carried out. No significant appearance changes were observed. The swelling behavior of the fuel pellets was negligible for all fuels. Axial and radial distributions of typical fission products (FPs) were analyzed by γ scanning. Cs moved partly to the gaps between the pellets and to the insulators. The distribution of ^{134}Cs was different from that of ^{137}Cs, especially for corundum-based fuel. The single phase fuel of U-YSZ showed great retention of ^{137}Cs.

PACS: 25.85.Ec; 28.41.B

1. Introduction

The concept of the rock-like oxide (ROX) fuel has been developed for the annihilation of excess plutonium in light water reactors (LWRs). Features of the ROX–LWR system are almost complete burning of plutonium and the direct disposal of the spent ROX fuels without reprocessing [1,2]. The ROX fuel consists of such phases as fluorite (yttria-stabilized zirconia; YSZ), $MgAl_2O_4$ (spinel), and Al_2O_3 (corundum). Plutonium, other actinides, and lanthanide fission products (FPs) solidify into the fluorite phase. The roles of spinel or corundum are to immobilize the alkali and alkaline earth nuclides and to improve thermal conductivity. The resistance of spinel to neutron irradiation is high [3], but may not be sufficient for fission fragments. For the improvement of swelling behavior, a particle-dispersed type fuel has been proposed [2,4].

In previous studies, in-pile irradiation examination of five ROX fuels was carried out [2,4–6]. These included a single phase fuel of a YSZ containing UO_2(U-YSZ), two particle-dispersed fuels of U-YSZ particles in spinel or corundum matrix, and two homogeneously-blended fuels of U-YSZ and spinel or corundum. It was confirmed that the particle-dispersed fuels showed lower swelling behavior than the homogeneously-blended fuels.

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doi:10.1016/j.jnucmat.2006.03.002

The regions suffering irradiation damage were observed as a thin layer of about 10 μm thickness on the surfaces of the YSZ particles. For spinel-based fuel, decomposition of the spinel and vaporization of MgO was observed. The spinel-based fuel showed high fission gas release compared with corundum-based fuel. The restructuring might be the primary cause for the high fission gas release. It was expected that the vaporization of MgO could be avoided by lowering the fuel temperature below 1700 K. On the other hand, under irradiation conditions of sufficiently high irradiation temperature to effect annealing of the amorphous corundum, no significant appearance changes such as swelling and heterogeneity of the fuel structures were observed for the corundum-based fuels.

We have performed a new irradiation examination of the ROX fuel with more appropriate conditions in order to confirm the irradiation behavior of ROX fuel. It was important to confirm the irradiation behavior of spinel-based fuel under the lower irradiation temperature, which is necessary to avoid the spinel decomposition. It is also essential for the evaluation of the behavior of corundum-based fuel to ascertain the occurrence of amorphization and subsequent swelling by lowering the irradiation temperature. The maximum center temperature of the fuel pellets was designed around 1500 K for spinel and corundum-based fuels. Furthermore, in previous experiments, it was difficult to compare the irradiation behavior of U-YSZ single phase fuel with that of other fuels because of much lower irradiation temperatures and burn-up caused by lower fissile densities and lower neutron flux. In this experiment, the fuels were designed to make the number density of fissile equal in all fuels in order to compare the irradiation behavior. The designed maximum center temperature of U-YSZ single phase fuel pellets was near 2500 K because of its low thermal conductivity.

For the post-irradiation examinations, visual inspection, profilometry, and γ scanning tests were carried out. The axial and radial distributions of typical FPs were obtained by γ scanning. The results of the examinations will be described and discussed.

2. Experimental

2.1. Preparation of fuels

Three fuels were prepared using 20% enriched U instead of Pu; a single phase fuel of U-YSZ (Z fuel) and two particle-dispersed fuels of U-YSZ particle in spinel or corundum matrix (S, C fuels). The compositions of these fuels are shown in Table 1.

The particle-dispersed type fuel was fabricated by sintering a mixture of U-YSZ particles and fine powder of matrix. The U-YSZ particles were prepared by crashing presintered U-YSZ pellets whose density was 65% of theoretical density (TD) and then sieving them. The particles of size 125–250 μm were sifted out. The shrinkage rate of the calcined particles and matrix at the sintering temperature was controlled to the same grade by adjusting the density of calcined particles. The U-YSZ particles were blended with fine powder of spinel or corundum and pressed to pellets. In all cases, the sintering of the pellet was carried out at 1820 K for 24 h in a stream of 4% H_2/He mixed gas. The size of each pellet was about 5.2 mm in diameter and 5.7 mm in height. The densities were about 93%TD for all fuel type pellets. Fig. 1 shows the appearance and the microscopic structure of the fabricated ROX fuel pellets. The homogeneous distribution of the U-YSZ particles in the matrix was confirmed by ceramography. The U-YSZ particle sizes were about 100–200 μm.

2.2. Irradiation

The irradiation capsule was designed to remove the heat of the fuels efficiently by cooling water and the amount of fissile elements was determined by burn-up calculations to control the irradiation temperature. Ten pellets of each fuel were loaded

Table 1
Fuel compositions

Fuel	Fuel type	Composition/mol%				^{235}U at. density/cm^{-3}
		YSZ[a]	UO_2	$MgAl_2O_4$	Al_2O_3	
Z	Solid solution	78.0	22.0	–	–	1.158×10^{21}
S	Particle-dispersed	33.35	29.65	37.00	–	1.159×10^{21}
C	Particle-dispersed	27.70	24.63	–	47.67	1.159×10^{21}

[a] YSZ: 78.6 mol%ZrO_2 + 21.4 mol%$YO_{1.5}$.

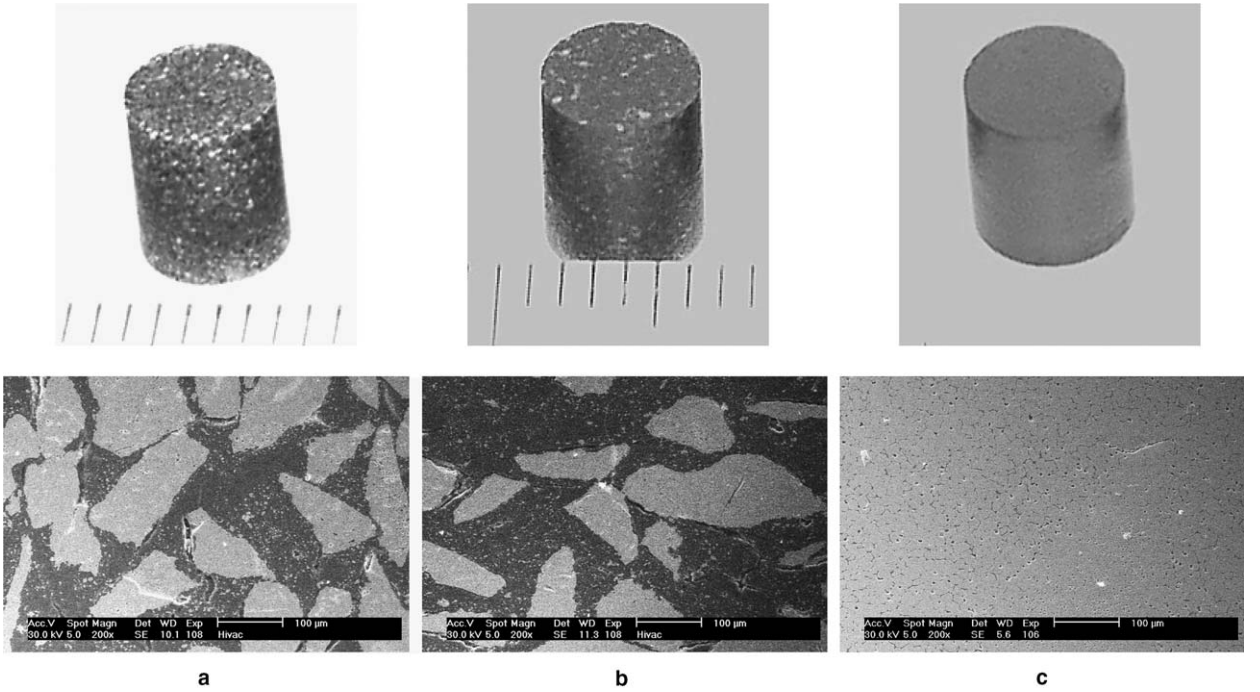

Fig. 1. Appearance and microstructure of the pre-irradiated fuel pellet of (a) S [U-YSZ + $MgAl_2O_4$, particle-dispersed type], (b) C [U-YSZ+Al_2O_3, particle-dispersed type], and (c) Z [single phase fuel of U-YSZ].

in a stainless steel cladding tube under He gas at ambient pressure and sealed off. Spinel pellets were used as insulators. The sizes of the fuel pins were 6.5 mm in diameter and 125 mm in length. The initial gaps between fuel pellet and cladding were about 130–180 μm.

These fuels were irradiated in the Japan Research Reactor No. 3 for about 280 days. The burn-ups measured by ^{148}Nd analytical method were 10.80, 10.65, and 11.96 FIMA% for S, C, and Z fuel, respectively, corresponding to about 25.7, 25.3, and 28.4 MW d kg^{-1} of LWR UO_2 fuel. The fuels were designed for maximum temperatures of about 1500 K for S and C fuels, and about 2500 K for Z fuel. The roughly estimated maximum linear power was 45 kW m^{-1}. The practical irradiation temperatures will be calculated in detail after the estimation of linear power by fluence monitor analysis is finished.

2.3. Post-irradiation examination

Non-destructive and destructive post-irradiation examinations were carried out at the Reactor Fuel Examination Facility in JAERI. The irradiation capsule was disassembled and three fuel pins were taken out.

Visual inspection of the fuel pins was carried out. The precise diameters of the fuel pins were measured from 4 directions at 45° intervals using an electronic micrometer. The appearance of the fuel pellets and the stack length was observed by X-ray radiography.

Axial distributions of typical FPs were analyzed by γ scanning from the plenum to the bottom of the fuel pin. Gamma rays from FPs such as ^{95}Zr, $^{106}Ru(^{106}Rh)$, ^{134}Cs,^{137}Cs, and $^{144}Ce(^{144}Pr)$ were detected through a thin slit placed in front of a germanium detector.

The fuel pins were cut-off and some pellets were taken out from fuel pins. The appearance of the fuel pellets was observed. Epoxy resin was injected into the remaining pellets within the cladding tube. After polishing the surface of the pellets, radial distributions of FPs in the fuel pellets were observed by micro-gamma scanning. Gamma rays were measured at intervals of 0.5 mm using a collimator with a 1 mm diameter.

3. Results and discussion

3.1. Non-destructive examination

Though the stainless steel cladding surface where fuel pellets were loaded was slightly discolored by

oxidation, no significant appearance changes were observed for all fuel pins. Any change in axial pellet stack length was not recognized from the comparison of X-ray radiography taken before and after irradiation. Diameter changes that exceed the experimental error were not observed by profilometry. It is suggested that the fuel pellet swelling was less than the initial gap for all the fuel types. The gaps between pellets were also confirmed from X-ray radiographs. The obvious inter-pellet gaps of C and S fuels were observed. The gaps of the Z fuel were not so clear, but sufficiently detectable. From these facts, the swelling behavior of fuel pellets was concluded to be negligible for all fuels.

The fuel pellets were removed from the fuel pins without bonding. In spite of crack formation, many of the C and S fuel pellets kept the original shape. In contrast, the Z fuel pellets were fragmented into about 20 pieces each.

The cross sectional appearances are shown in Fig. 2. Some radial cracks formed in the S fuel. Although the center region of the S fuel pellet seemed to be discolored, the decomposition of spinel and subsequent restructuring that occurred in previous experiments at high irradiation temperature (~2000 K at the center of pellets) were not observed [5,6]. A central hole was not formed either. The number of cracks of C fuel was least among the three fuels. The gaps between the pellets and the cladding were clearly observed for the C fuel. In the Z fuel pellets, several radial cracks and a central hole within 0.8 mm diameter can be observed, as is similarly observed in high burn-up LWR UO_2 fuel. Probably, the central hole was caused by densification and migration of pores at the high temperature. The Z fuel pellet was very porous in the middle region and the growth of columnar grains was observed from middle towered center region.

The appearance changes of particle-dispersed fuels seemed to be smaller than Z fuel.

3.2. Fission product (FP) distributions

The axial distributions of ^{95}Zr, ^{137}Cs, and ^{134}Cs together with X-ray radiographs of the fuel pins are shown in Fig. 3. The dents of the distributions of ^{95}Zr coincided well with pellet–pellet gaps for all fuels. It showed that nonvolatile Zr existed only in the fuel pellets. On the other hand, a small amount of Cs moved to the gaps between the pellets and to the insulators. The estimated amounts of ^{137}Cs that released from the fuel pellets were about 5, 2, and below 1% for C, S, and Z fuel, respectively.

The ^{134}Cs and ^{137}Cs showed different distributions, particularly for the C fuel (Fig. 3(b)). Although there were only small amounts of ^{137}Cs, it is clearly evident that ^{134}Cs exists in plenum region for all fuels. The estimated released amounts of ^{134}Cs were about 24%, 6%, and 3% for C, S, and Z fuel, respectively. The behavior is probably due to differences in the production processes between ^{134}Cs and ^{137}Cs. Almost all ^{137}Cs is produced by fission directly and the half-lives of the parent nuclides are short. On the other hand, much of the ^{134}Cs comes from neutron capture of ^{133}Cs, generated by the decay of ^{133}Xe whose half-life is 5.25 days. The half-life of ^{133}Xe might be long enough to behave as a gas. In other words, the distribution of ^{137}Cs showed only the behavior of Cs (alkali metal), while the distribution of ^{134}Cs showed not only behavior of Cs, but

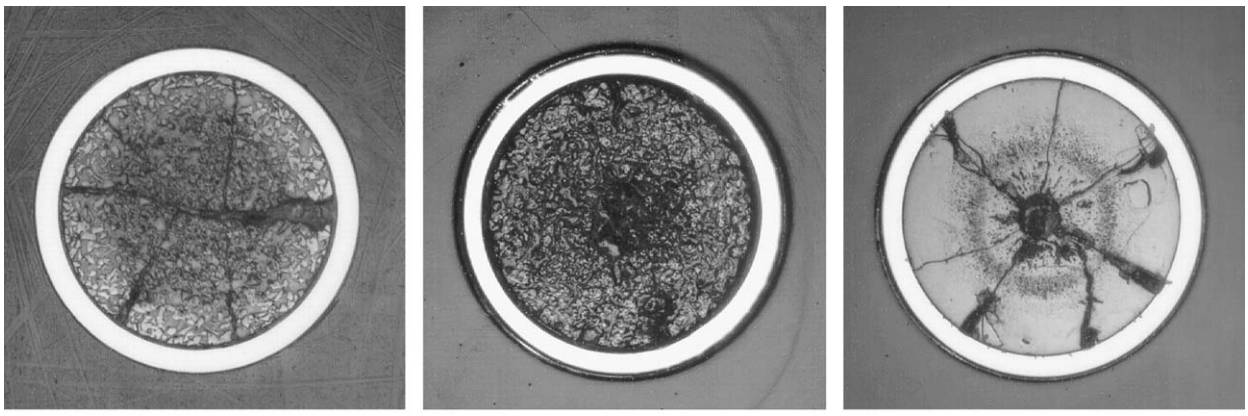

Fig. 2. Cross sectional views of irradiated fuel pellets. (a) S, (b) C, and (c) Z fuels.

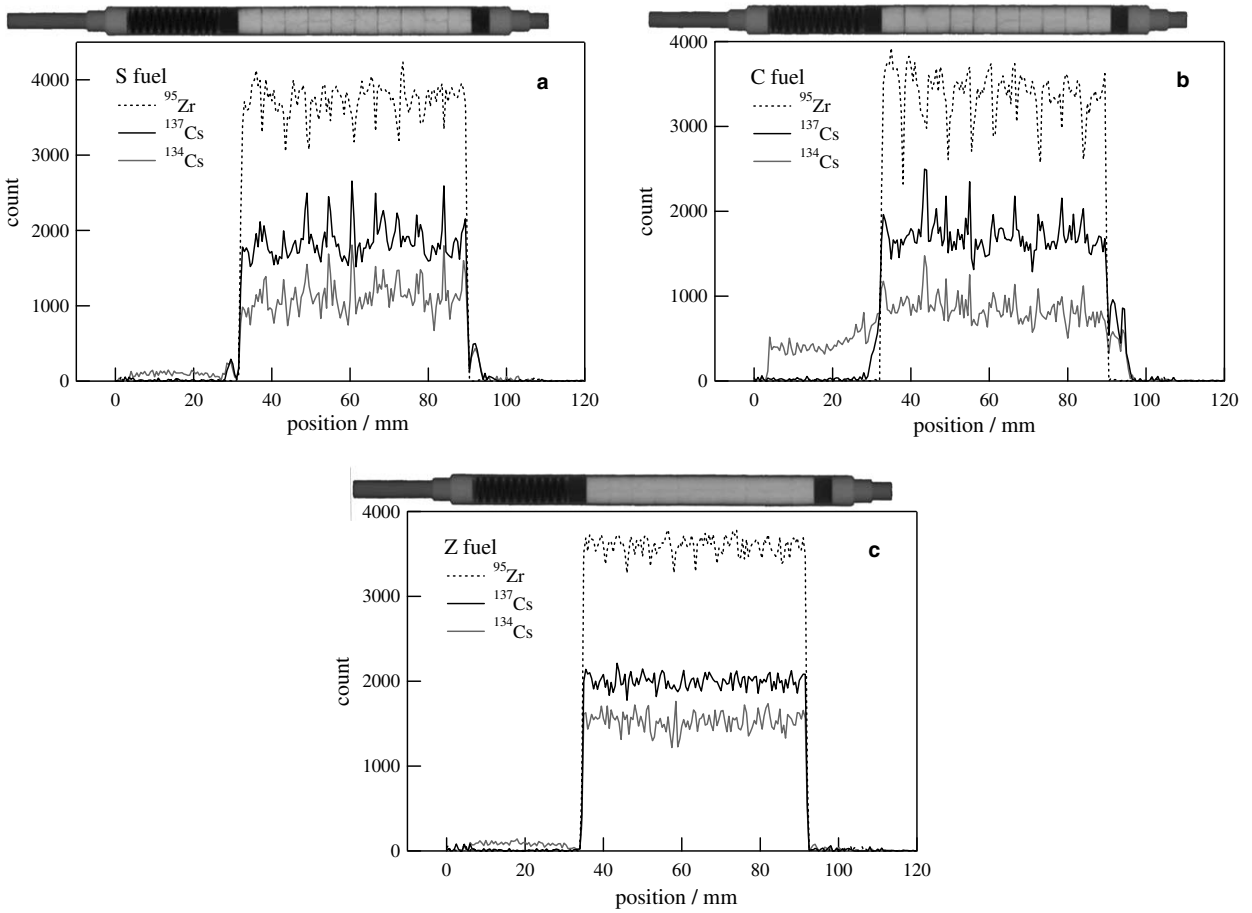

Fig. 3. X-ray radiography and γ scanning of ^{95}Zr, ^{137}Cs, and ^{134}Cs for (a) S, (b) C, and (c) Z fuel pins.

also reflected the behavior of Xe (gas). From the distribution of ^{134}Cs, it is expected that large amounts of FP gases were released from the fuel pellets for the C fuel. The corundum did not cause the swelling of the pellet, but it appears the retention ability for Cs and FP gas is very low. The spinel, which was not decomposed, showed higher Cs retention ability than corundum. Probably, the gas retention ability of S fuel is also higher than that of C fuel. In previous tests, the fuels also showed different distributions between ^{134}Cs and ^{137}Cs, but the difference was unclear, due to the lower yields of ^{134}Cs at lower burn-up.

Tomographic images of ^{95}Zr and ^{137}Cs for each fuel pellet type are shown in Fig. 4. Nonvolatile FPs such as ^{95}Zr and ^{106}Ru showed uniform distribution throughout the pellet surface, except for the Z fuel. The amount of ^{95}Zr in the Z fuel seems to decrease in the center region (Fig. 4(c)) though not caused by migration of Zr from the center to periphery, but due to the central hole. After correcting for the area of the central hole, it was confirmed that the decrease came from the reduction of the effective area by the central hole. On the other hand, much of the Cs escaped from center region and showed uneven distribution. The unevenness of the Cs distribution for the C fuel was the largest among the fuels. Due to the effects of wet polishing, it was not possible to ascertain the deposition point of Cs by comparing the distribution to the cross sectional appearance.

Radial distribution plots for ^{137}Cs are shown in Fig. 5. Each data point represents the circular average. From the viewpoint of thermal conductivity, it is expected that the temperature of the Z fuel pellets was highest among the three fuels. However, the amount of ^{137}Cs that escaped from the fuels decreases in the order C, S, and Z. Nevertheless,

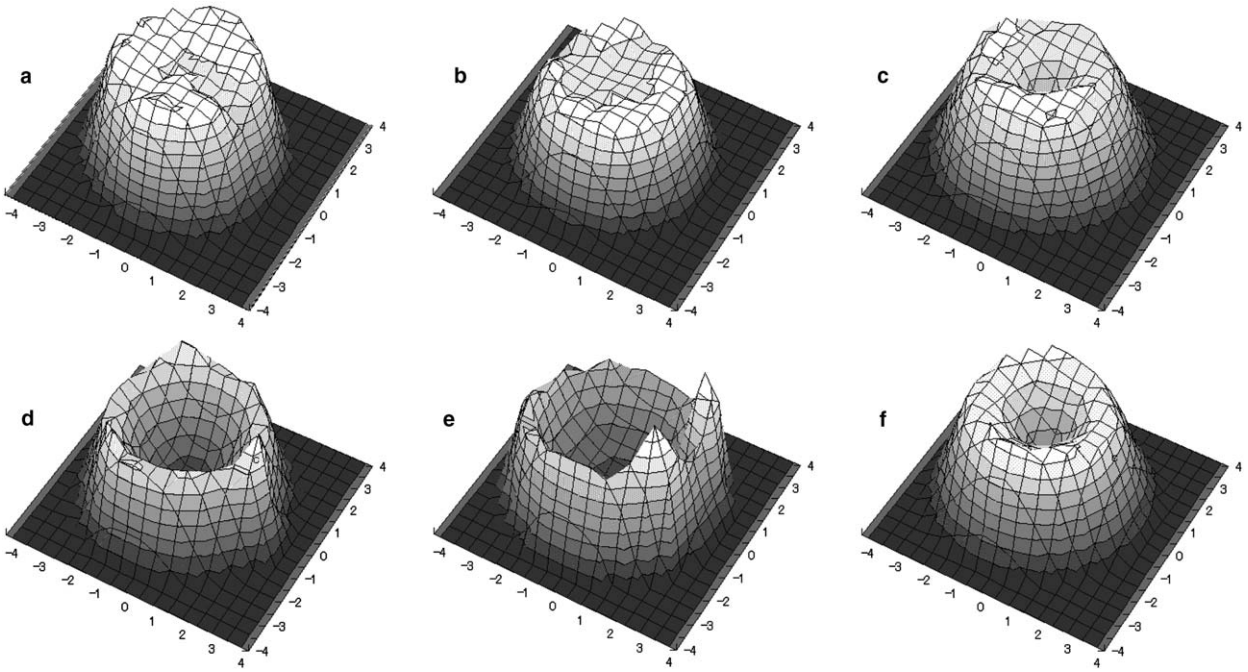

Fig. 4. Tomographic images of ^{95}Zr for (a) S, (b) C, and (c) Z fuels and those of ^{137}Cs for (d) S, (e) C, and (f) Z fuels.

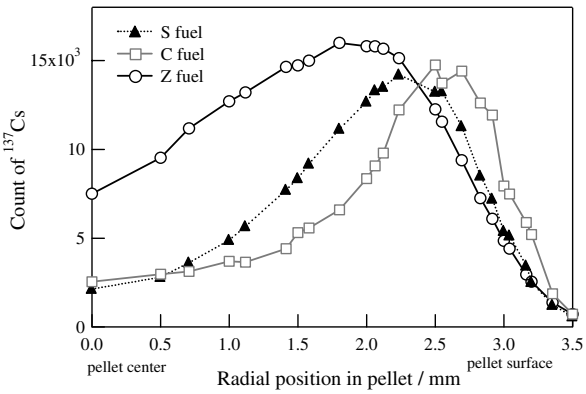

Fig. 5. Radial distribution of ^{137}Cs along a line from pellet center to the surface for S, C, and Z fuels.

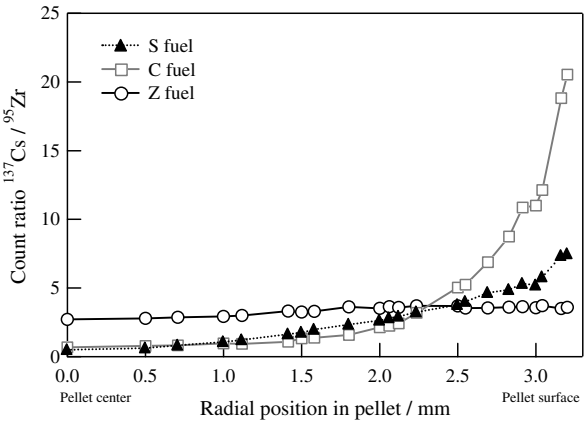

Fig. 6. Ratio of $^{137}Cs/^{95}Zr$ along a line from pellet center to the surface for S, C, and Z fuels.

comparing the distributions of the S and Z fuels, the distributions in the periphery region were similar and the concentration of ^{137}Cs in the S fuel decreased in the center region. It showed that the much of the ^{137}Cs escaped from the center of the fuel pellet. On the other hand, for the C fuel, the concentration of ^{137}Cs shifted to the pellet surface region, while decreased markedly toward the fuel center. The ^{137}Cs escaped from the center and migrated to the periphery.

Fig. 6 shows a radial plot of the ^{137}Cs to ^{95}Zr ratio along a line from the pellet center to the surface. Whereas there was a significant loss of Cs from the central region and migration to the periphery of C fuel, the Z fuel showed a very high degree of retention. The degree of retention for the S fuel was higher than that for the C fuel.

The irradiation behaviors were entirely different between the two particle-dispersed type fuels with respect to the axial distributions of ^{134}Cs and the

behavior of ^{137}Cs in this experiment. Since the U-YSZ particles for the S and C fuels were similar, it is considered that the time needed to diffuse the gases and Cs to the boundary of the U-YSZ particles were almost the same between the S and C fuels. The difference in behavior was caused by the difference in the properties of the matrices. It is expected that the amorphization of corundum and/or the increase in the fuel center temperature accompanied by a reduction in the thermal conductivity with the amorphization caused the difference.

4. Summary and conclusions

Three ROX fuels using 20% enriched U as a fissile were prepared and irradiated. No significant appearance changes such as swelling and fragmentation were observed for all fuel pins and pellets. Although several radial cracks were formed, no restructuring and no central hole were observed in spinel and corundum-based fuel pellets. On the other hand, a central hole and a porous structure were observed in the U-YSZ single phase fuel pellet.

Axial and radial distributions of FPs were observed by γ scanning. ^{134}Cs and ^{137}Cs showed different distributions especially for the corundum-based fuel. This was caused by the difference in the decay series between ^{134}Cs and ^{137}Cs. It is expected that many FP gases were released from the corundum-based fuel pellets according to the distribution of ^{134}Cs.

The irradiation behavior of spinel-based fuels, which were irradiated under the avoidable condition of spinel decomposition, was superior to corundum-based fuels, for their retention of Cs and FP gases. The temperature of U-YSZ single phase fuel pellets was highest among the three fuels, because of its low thermal conductivity. Nevertheless the U-YSZ single phase fuel showed the greatest retention of ^{137}Cs.

Acknowledgements

The authors wish to acknowledge Mrs S. Miyata and Y. Kawasaki for burn-up measurement by ^{148}Nd analytical method for the irradiated ROX fuels.

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