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Helium release from the uranium–plutonium mixed oxide (MOX) fuel irradiated to high burn-up in a fast breeder reactor (FBR)

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ABSTRACT

The helium releases were investigated in FBR fuel pins irradiated to high burn-up. The released amounts increased with the increase of burn-up, but no burn-up dependence of their release fraction to total amounts generated was seen. The contents of ^{241}Am in fuel pellets affected the released amounts. The effect of released helium on the internal pressure in fuel pins was smaller in FBR fuel pins than in LWR fuel pins, but released helium in fuel pins containing increased amounts of ^{241}Am, which are anticipated in future FBRs, is expected to affect the internal pressure.

1. Introduction

Helium is generated and built up in uranium–plutonium mixed oxide (MOX) fuel by the ternary fission of uranium and plutonium, the nuclear reaction of oxygen with a neutron, $^{16}O(n, \alpha)^{13}C$ and the α decay of actinide elements. Some of these helium releases are into the fuel-cladding gap and plenum of the fuel pins, and this helium affects the internal pressure and thermal conductance across the fuel-cladding gap. Thus, it is very important to know the helium release behavior from the MOX fuel matrix from the viewpoint of fuel pin integrity.

Some studies have been published on helium release from oxide fuels. Initially these studies theoretically investigated the solution energy and migration behavior based on models of an atomistic mechanism [1,2]. Several of the studies were carried out on light water reactor (LWR) fuels [3–5]. Ronchi and Hiernaut [6] took tiny specimens from uranium oxide (UO_2) and MOX fuels irradiated to high burn-up in an LWR, and then heated them in a Knudsen cell equipped with a mass spectrometer and obtained the diffusion coefficient. On the other hand, Yang and Crowther [7] collected the data on helium releases from UO_2 fuels irradiated in commercial reactors and analyzed these data as a function of burn-up.

In order to develop fast breeder reactor (FBR) fuel, a number of irradiation tests of MOX fuels have been carried out in the range of burn-up to 110 GWd/t using the experimental fast reactor JOYO. As one of the post irradiation examinations (PIEs) on these irradiated MOX fuels, the amount of helium released from the fuel matrix has been measured by pin puncture tests.

In the present study, the data on helium release obtained in the above irradiation campaigns were analyzed as a function of burn-up. The total amounts of helium generated in the fuel matrix were theoretically calculated and then the release fractions were determined. The results obtained were compared with those of other researchers [3,7]. Furthermore, these data were compared with data on fission gas release and the effects on the fuel performance were discussed.

2. Experimental

2.1. Specimens and irradiation conditions

Helium releases were measured in the fuel pins of 17 fuel assemblies irradiated to different burn-ups in JOYO. Sixteen assemblies were driver fuel assemblies for the JOYO core and one was the type C assembly developed for irradiation test to high burn-up. Fig. 1 illustrates the fuel pin configuration in both types of fuel assemblies. Detailed explanations were given in Ref. [8].

After dismantling these fuel assemblies, 133 fuel pins were subjected to puncture tests. Table 1 shows the numbers of these fuel pins in each fuel assembly, together with key specifications of fuel pellets and irradiation conditions of the fuel assemblies. The fuel pellets had Am contents ranging from 0.035 to 0.273 wt.% per metal of MOX fuel. Their densities ranged from 85% to 94% of the theoretical density, and their O/M ratios ranged from 1.94 to 1.98. Average burn-ups and linear heat ratings (LHRs) were from 26 to 110 GWd/t and 139 to 317 W/cm, respectively.

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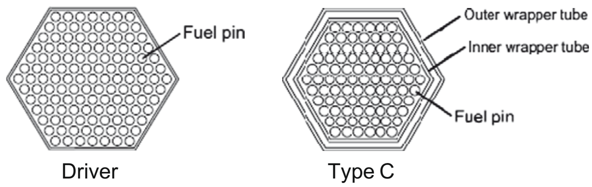

Fig. 1. Cross-sectional views of two fuel assemblies.

2.2. Measurement of helium release from fuel pins

Helium releases from the irradiated MOX fuel pins were measured by puncture tests. A schematic drawing of the puncture test setup is shown in Fig. 2. The volume of the measurement vessel was not so different from the known volume of the calibration vessel. First, both measurement and calibration vessels were evacuated. After closing valve A, nitrogen gas at a pressure of 1 atm was introduced into measurement vessel from the gas inlet. Then, valve B was closed and valve A was subsequently opened. The volume of the measurement vessel was determined from the difference of the pressures before and after opening valve A. After closing valve A and evacuating the measurement vessel again, the fuel pin was punctured by a laser beam and the gas in the fuel pin spread into the measurement vessel. Next, a part of the gas was flowed into the measurement system installed outside of the hot cell by opening valve B. Its composition was analyzed with a gas chromatograph.

The free volume in the fuel pin was measured as follows. First, the measurement vessel and the fuel pin were evacuated after finishing the above measurement. Secondly, the nitrogen gas at a pressure of 1 atm was introduced into both the measurement vessel and the interior of the fuel pin. Thirdly, the puncture pin hole was sealed by heating the pin with the laser beam. Fourthly, the

Table 1
Key specifications of fuel pellets and irradiation conditions of fuel assemblies.

S/A	Type of SA	Number of pins	Fuel weight (g)	Am content (wt.%)	Density (% TD)	O/M	Pin averaged burn-up[a] (GWd/t)	Pin averaged LHR[b] (W/cm)
PFD210	Driver	2	96	0.164	93	1.94	44–47	195–200
PFD209	Driver	3	95	0.167	94	1.97	58–60	239–249
PFD135	Driver	19	96	0.107	94	1.95	47–50	290–307
PFD153	Driver	7	96	0.037	93	1.98	65–71	249–270
PFD207	Driver	15	94	0.166	92	1.98	42–56	139–191
PFD139	Driver	5	97	0.036	94	1.95	40–42	292–312
PFD254	Driver	16	96	0.273	93	1.98	39–48	185–233
PFD001	Driver	4	98	0.041	94	1.96	32–39	312–317
PFD018	Driver	4	97	0.035	93	1.98	39–44	247–278
PFD029	Driver	4	97	0.041	93	1.97	41–48	227–262
PFD035	Driver	14	96	0.035	93	1.97	26–32	176–219
PFD036	Driver	3	97	0.040	94	1.96	35–45	169–215
PFD105	Driver	6	95	0.038	93	1.97	48–51	293–316
PFD068	Driver	1	95	0.051	93	1.98	41	301
PFD304	Driver	12	98	0.170	94	1.98	61–66	239–260
PFD310	Driver	6	98	0.267	94	1.98	49–59	158–207
PFC030	Type C	12	120	0.191	85	1.95	99–110	253–289

[a] Burn-up (GWd/t): thermal output per unit weight of fuel pellet.
[b] Linear heat rating (W/cm): thermal output per unit length of the fuel pin.

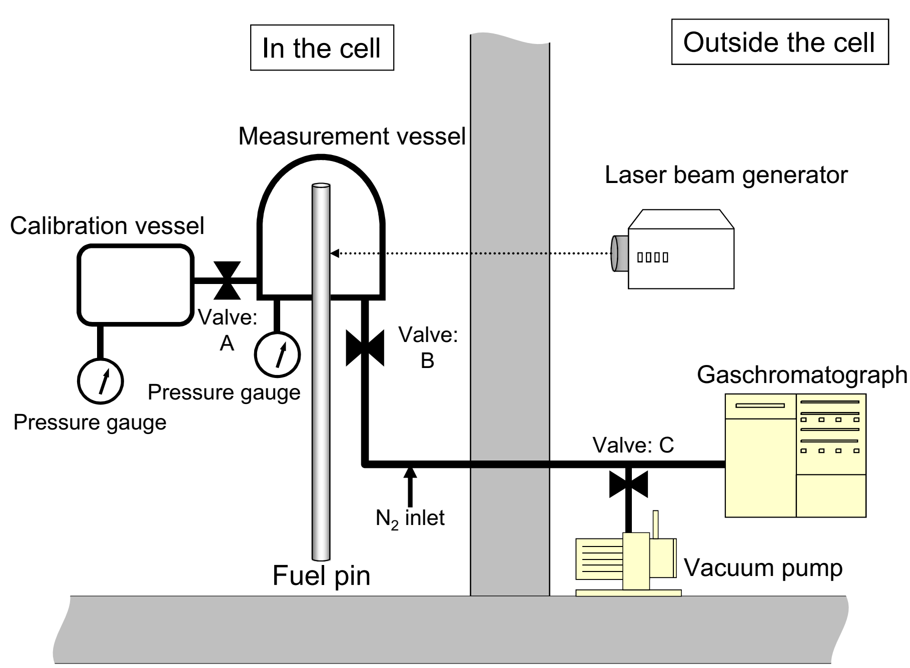

Fig. 2. A systematic diagram of puncture test setup.

measurement vessel was evacuated. Finally, the fuel pin was punctured by the laser beam after closing valve B. The volume of free space in the fuel pin was determined from the difference before and after puncturing. The total amount of helium was calculated, using the volume of measurement vessel, free volume in the fuel pin, gas composition, the pressure at the time of pin puncture and temperature. The amount of helium released from the fuel pellet was determined by subtracting the amount of filled helium at the time of fuel pin fabrication from the total amount of helium.

The accuracy of the pressure gauge was ±0.08%. In this pin puncture test, less than 10 gas pressure measurements were made. Then, the total gas volume was measured within an error of ±1.32%. However the amount of helium released from the fuel pin was measured within ±5% including the error of composition analysis due to the gas chromatograph use.

3. Theoretical prediction of helium amount generated in the fuel matrix

In a fuel pellet, helium is generated by ternary fission of heavy nuclides, the reaction of oxygen with a neutron, $^{16}O(n, \alpha)^{13}C$ and α decay. The helium generation by ternary fission was calculated using the fission yield reported by Rider [9], as follows:

$$Q(\mathrm{He}) = \sum \sigma_i \times \phi \times N_i \times Y_i \times t$$

where $Q(\mathrm{He})$ is the amount of helium generation, σ_i is the fission cross section of fissile nuclide i, ϕ is neutron flux, and N_i is the number of fissile nuclide i, Y_i is the yield of helium, and t is irradiation time.

The helium generations by both α decay and the (n, α) reaction were calculated using the ORIGEN-2 code. In these calculations, the operation history and nuclide composition of as-fabricated fuel were taken into consideration. In addition, neutron flux in the reactor was calculated by the MAGI code developed for the calculation of neutron flux in JOYO [10]. This neutron flux depended on the axial position of a fuel pin, but it was averaged along the axial direction and it was assumed that this averaged value was constant along the axial direction.

4. Results and discussion

Fig. 3 shows the amounts of helium released from the fuel matrix as a function of burn-up. The released amounts increase with the increase of burn-up and the data above the burn-up of nearly 40 GWd/t are scattered. In order to clarify the reason for this, attention was paid to the content of ^{241}Am in as-fabricated fuel because this nuclide generates ^{242}Cm which undergoes α decay with a short half life.

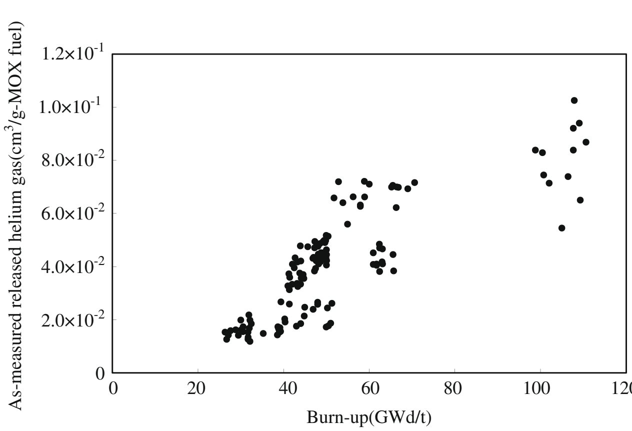

Fig. 3. Helium release as a function of burn-up.

Fig. 4 shows the amounts of helium released from the fuel matrix as a function of ^{241}Am content at burn-ups from 50 to 60 GWd/t. It should be noted that the released amounts increase with the increase of ^{241}Am content. In order to theoretically confirm this effect, the amounts of helium, which are generated by three mechanisms, α decay, ternary fission and the (n, α) reaction, were calculated by the ORIGEN-2 code in the pellets containing 0.038 wt.% and 0.267 wt.% of ^{241}Am as an example. As shown in Fig. 5, the amount of helium generated by α decay is larger in the pellet containing 0.267 wt.% of ^{241}Am than in the one containing 0.038 wt.% of ^{241}Am, although the amounts generated by two other mechanisms is nearly same in both pellet types. The differences of average measured amounts of helium generated by α decay between both types is about 0.03 cm^3/g-MOX fuel and is nearly equal to the theoretical value shown in Fig. 5. From this result, the content of ^{241}Am is judged to have a large effect on the amounts of helium released.

Fig. 6 compares measured release data with the amounts generated in the fuel matrix which were theoretically calculated by the method described in Section 3. Most of the measured release data are found in the range from 50% to 100% of calculated generation amounts, although they are widely scattered. In addition, the deviation of the measured release amounts increases with the increase of release amount. This tendency was previously seen in the data reported by Bairiot and Lippens [1].

The helium release fractions (%) obtained from these data are shown in Fig. 7 as functions of burn-up and linear heat rating. No

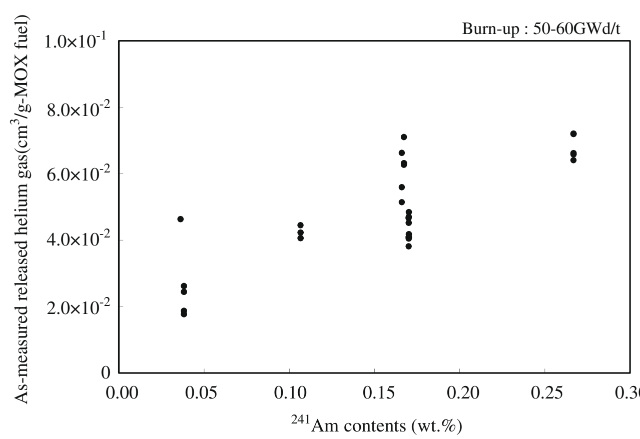

Fig. 4. Helium release as a function of ^{241}Am contents.

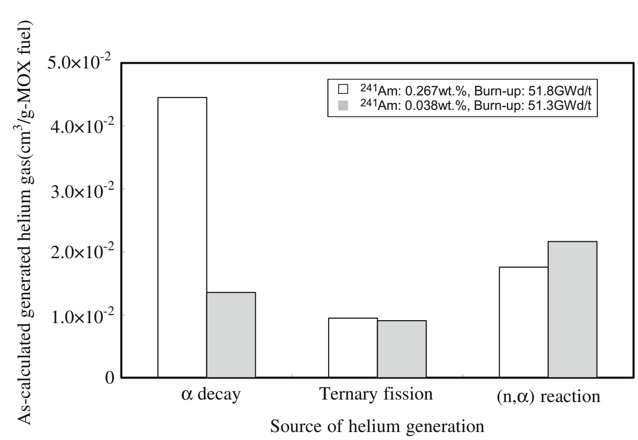

Fig. 5. Comparison of helium generation by three mechanisms.

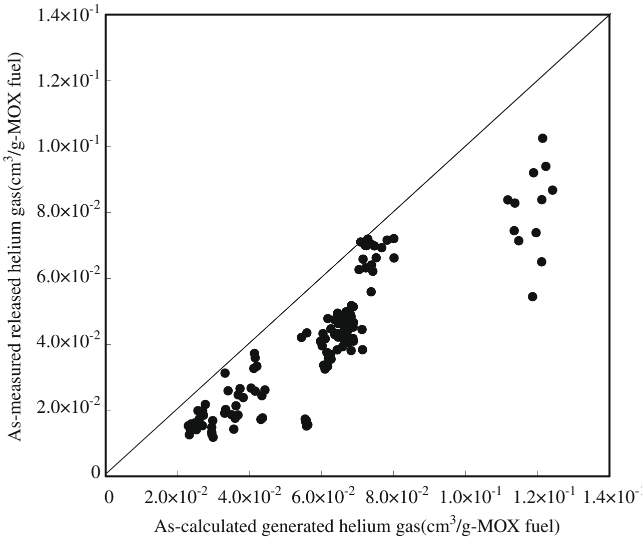

Fig. 6. Comparison of measured data with the theoretically calculated generated amounts.

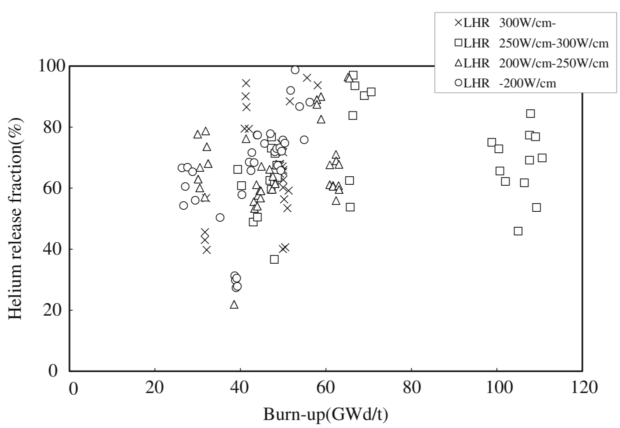

Fig. 7. Helium release fractions (%) as functions of burn-up.

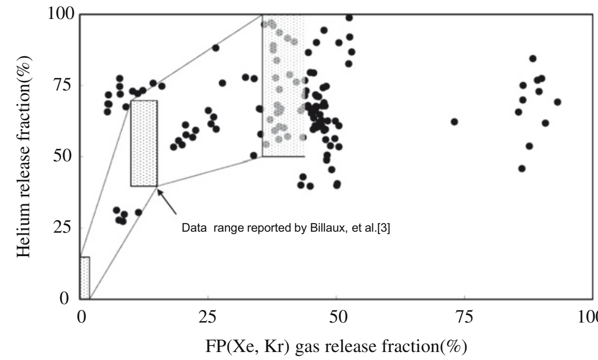

Fig. 8. The relationship between both fractions of fission gas and helium releases.

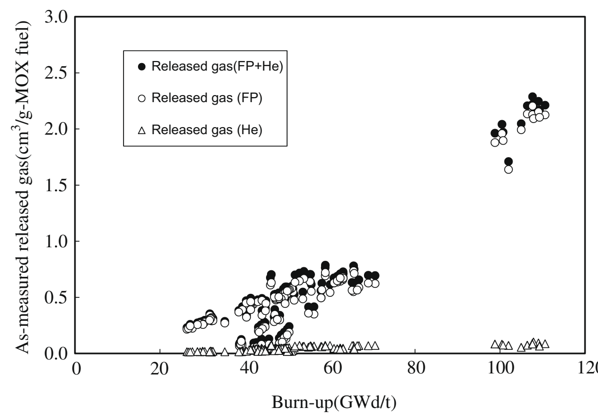

Fig. 9. The comparison between both amounts of fission gas and helium release.

noticeable dependencies of helium release fractions on the burn-up and linear heat rating can be seen. The relationship between both fractions of fission gas and helium release is shown in Fig. 8. Helium release fractions are larger than fission gas release fractions in the region where the fission gas release fraction is lower than 50%, but they become slightly smaller beyond this region. In addition, the relationship between fission gas release and helium release fractions in the range of low release fractions shows a good agreement with the data reported by Billaux et al. [3].

As an explanation of the tendencies shown in Figs. 7 and 8, the following is suggested. Based on the theory [11] developed for the fission gas release, the noble gas atoms (helium, krypton and xenon) generated in the oxide fuel diffuse to the grain boundary, through repeated processes of trapping into and resolution from the lattice defects in the fuel matrix. These gas atoms accumulated on the grain boundary grow into gas bubbles and then some of the bubbles become connected. The connectivity of these gas bubbles increases on increasing the noble gas amounts with burn-up. When the amount of noble gas increases further with the increase of burn-up and reaches a threshold value, these gas bubbles will form gas tunnels on the grain boundary surfaces and edges, which reach the free space in the fuel pin where the noble gas atoms are released. Recently, Tikare, et al. [12] have theoretically described this fission gas behavior, using a simulation based on the Potts kinetic Monte Carlo (kMC) model.

The atomic radius of helium is smaller than the atomic radii of the fission gas atoms (krypton and xenon). From this fact, the helium gas atoms should easily diffuse in the fuel matrix, and the times are expected to be short for the migration of gas atoms to the grain boundary surfaces and edges, leading to the formations of bubbles and tunnels. In this study, the helium releases were measured in the range of burn-up above 20 GWd/t. If the threshold burn-up, i.e. the point when helium release begins through tunnels formed on the grain boundary surfaces and edges, is less than 20 GWd/t, almost all helium will be released at burn-ups above 20 GWd/t. Thus, the burn-up dependence of helium release shown in Fig. 7 and the higher helium release fraction shown in Fig. 8 are understood.

The released helium affects the internal gas pressure and thermal conduction in the fuel pin. Fig. 9 shows the amounts of fission gas and helium releases as a function of burn-up. The effect of released helium on the overall internal pressure is small compared with the effect of released fission gas. However, the ^{241}Am content affects the helium pressure as shown in Figs. 4 and 5. When FBR development progresses, it is expected that MOX fuels containing large amounts of ^{241}Am will be utilized as core fuels. Based on predictions by the ORIGEN-2 code, the helium pressure represents about 20% of the pressure due to all fission gases if the ^{241}Am content in MOX fuel is 5 wt.%.

5. Conclusion

Helium releases were studied as a function of burn-up in a large number of FBR MOX fuel pins irradiated to high burn-up. The released amounts of helium increased with the increase of burn-up, but their data were scattered in the region of high burn-up. This was understood to be caused by the differences of ^{241}Am contents among fuel pellets, because this nuclide generates ^{242}Cm which undergoes α decay with a short half life.

Theoretical amounts of helium which were generated in the fuel pellets by ternary fission, the $^{16}O(n, \alpha)^{13}C$ reaction and α decays of actinide nuclides, were calculated and release fractions were determined. No clear dependence of these fractions on the burn-up was seen in the fuel pins irradiated above 20 GWd/t. The effect of released helium on the internal pressure of the fuel pins was smaller in the FBR fuel pins than in LWR fuel pins, but this effect would become large in FBR fuel pins containing a large content of ^{241}Am, which are anticipated to be burned in future FBRs. The relationship between helium release and fission gas release fractions was in general agreement with the results for LWR fuel obtained by Billaux et al. [3]

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