Transactions of the Korean Nuclear Society Autumn Meeting
Yeosu, Korea, October 25-26, 2018

Qualitative Analysis of Fission Gas Release in the Micro-cell Fuel Pellets under Normal Operation Based on UO_2 Experience

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1. Introduction

After the Fukushima accident in March 2011 in Japan which spewed tremendous amount of radioactivity into the atmosphere [1], Korea Atomic Energy Research Institute (KAERI) has begun to develop micro-cell fuel pellets with an aim to reduce fission gas release (FGR) during accident conditions as well as under normal operations [2].

Since both irradiation experience and post-irradiation examination (PIE) data for the micro-cell pellets are either limited or unavailable, it is difficult at present to confirm quantitatively whether FGR in the micro-cell pellets would be reduced as intended, and if so, how much reduction in gas release would be achieved.

Based on UO_2 pellet experience accumulated for 60 years regarding FGR, qualitative analysis is made in this paper about how fission gas atoms would behave in the micro-cell pellets and what would be their FGR considering UO_2 pellet's gas release under normal operating conditions.

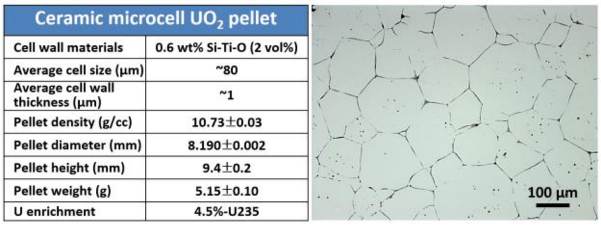
2. Micro-cell Fuel Pellets

KAERI is developing two types of micro-cell fuel pellets that consist of cells surrounded by wall material, where wall is intended for keeping fission gas atoms within each cell and thus preventing them from getting out of it, effectively reducing FGR significantly in the cell and ultimately in the micro-cell pellet itself [3]. Depending on the material used for making the wall, they are either ceramic micro-cell or metallic micro-cell pellet.

2.1 Microstructure of the Ceramic Micro-cell Pellet

The wall in the ceramic micro-cell pellet is composed of oxide phase with chemical affinity to volatile elements such as Cs and Iodine, so that the wall can capture them during reactor operation and also immobilize stable fission products (Xe and Kr) as well. In addition, the increased retention capability of the fission products will reduce stress corrosion cracking at the inner surface of the cladding as well as fuel rod internal pressure [2].

A representative fabrication data and microstructure of the ceramic micro-cell pellets used for Halden irradiation test within the framework of cooperation with ThorEnergy is shown in Fig. 1 [4]. It is to be noted that the ceramic micro-cell pellet is composed of a large UO_2 grain with the size of 80-120μm and the wall with the thickness of 1μm. Due to the decision made in June 2018 by IFE to shut down permanently the Halden reactor after 60 years of operation, the final burnup achieved in the ceramic micro-cell pellets was 16,000 MWd/kgU.



Ceramic microcell UO_2 pellet	
Cell wall materials	0.6 wt% Si-Ti-O (2 vol%)
Average cell size (μm)	~80
Average cell wall thickness (μm)	~1
Pellet density (g/cc)	10.73±0.03
Pellet diameter (mm)	8.190±0.002
Pellet height (mm)	9.4±0.2
Pellet weight (g)	5.15±0.10
U enrichment	4.5%-U235

Fig. 1. Fabrication data and microstructure of the ceramic micro-cell pellet for an irradiation test in the Halden reactor [4].

2.2 Microstructure of the Metallic Micro-cell Pellet

Mo and Cr were selected as wall materials mainly because they have relatively high melting temperature, high thermal conductivity, and manageable neutron absorption cross section. Above all, it is very important that Mo and Cr should be compatible with UO_2 matrix for the sintering conditions used for pellet fabrication [2]. The walls made by these metals would reduce gas release not only by blocking physically the movement of gas atoms and also through the reduced diffusivity of gas atoms caused by higher thermal conductivity.

Fig. 2 shows the microstructure of the metallic micro-cell pellets with 5vol% Cr as wall material [5]. The metallic micro-cell pellet consists of a large cell with the size of 300μm, which itself is composed of many small UO_2 grains of around 10μm, and its wall thickness is 2-7μm. These pellets, together with the ceramic micro-cell pellets described above, achieved the burnup of 16,000 MWd/kgU in the Halden reactor.

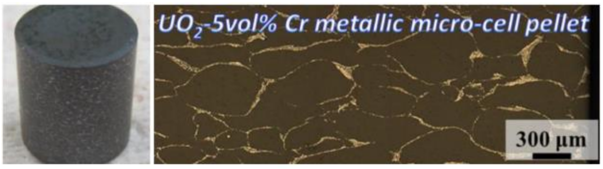


Fig. 2. Microstructure of a Cr metallic micro-cell pellet for an irradiation test in the Halden reactor [5].

3. FGR Mechanism in the Micro-cell Fuel Pellets

3.1 FGR Mechanism in the Conventional UO_2 Pellet

More than 40 billion UO_2 pellets have been irradiated since they were introduced into commercial nuclear reactors about sixty years ago. Hence very extensive studies had been and are still being made to explain how fission gas in the UO_2 pellets are released from UO_2 matrix to fuel outside through grain boundaries.

Fission gas release in the UO_2 fuel is summarized as follows [6]: fission gas atoms are produced in the matrix and migrate to the grain boundaries; bubbles nucleate, grow, and interconnect at the grain boundaries until they contact grain edges; gas moves along the interconnected grain edge tunnels until it reaches a free surface and released. Fig. 3 shows an illustration of each stage of gas release described above.

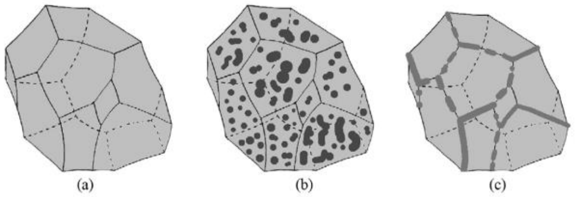


Fig. 3. Illustration of the process of gas release in UO_2 fuel: (a) gas production in the matrix and diffusion to grain boundaries; (b) formation and growth of bubbles at grain boundaries; (c) formation of release pathways by coalescence of bubbles at grain edges [6].

3.2 FGR Mechanism in the Ceramic Micro-cell Pellet

Gas atoms behavior in the ceramic micro-cell—cell interior except for the wall—would be the same as that in the matrix of conventional UO_2 pellet, because the cell interior is also the UO_2 matrix. So FGR in a specific ceramic micro-cell and then FGR in a whole ceramic micro-cell pellet composed of many ceramic cells would be determined by the role of the cell wall. For example, some issues for the cell wall to be resolved regarding gas release would be as follows: Would gas atoms diffuse into the wall from the UO_2 matrix? And, if so, would they form gas bubbles within the wall and then interconnect each other to make release pathways through which gas can be released to open space?

In the case of conventional UO_2 fuel pellet, grain boundaries are empty space where gas atoms can easily enter from the matrix and then form bubbles without much constraint, eventually making release pathways to the outside of the fuel pellet. The width of the grain boundary ranges from 5 to 10Å [7-8], two times the lattice constant (5.47Å) of UO_2 crystalline structure.

On the other hand, the cell wall in the ceramic micro-cell pellet is different from the grain boundary in two aspects. First, the wall is not an empty space but it is a ceramic material such as the mixture of SiO_2 and TiO_2, making it difficult for the gas atoms to form bubbles because void volume needed for bubble formation is unlikely to be available. Second, the width of 1μm as shown in Fig. 1 is at least 1000 times larger than the grain boundary width. So even if it is possible for the gas atoms to collide each other and form bubbles in the wall, given the big space in the wall available for the gas atoms, it would be practically difficult for them to do so.

This argument implies that, if the cell wall is intact during irradiation, the gas atoms produced in a specific cell would be almost impossible to escape from the cell due to the presence of the wall, leading to negligible gas release by diffusion except for direct release by recoil and knock-out within 5-10μm distance from free surface area. If this is the case, there would be two kinds of bubbles in the cell: small ones having diameter from one to 10nm with the density of 10^{23} m^{-3} [6] and large bubbles located next to the wall as shown in Fig. 4(a). The large bubbles would likely to be formed by the gas atoms that arrive at the wall by diffusion but are unable to diffuse into the wall due to the wall's resistance of incorporating them into its crystalline structure.

However, gas release could still occur in the ceramic micro-cell pellet by the diffusion of gas atoms through the area of the cells whose walls are partly unavailable as shown in Fig. 4(b).

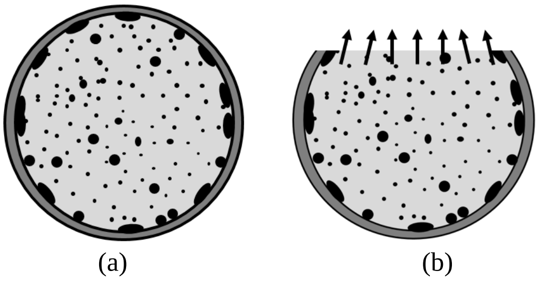


Fig. 4. (a) Gas bubbles that would likely to be observed in a ceramic micro-cell pellet with intact wall during irradiation; (b) Gas release in a ceramic micro-cell pellet in which cell wall does not exist in some region.

3.3 FGR Mechanism in the Metallic Micro-cell Pellet

The microstructures of the two micro-cell pellets are different: while each cell itself in a ceramic micro-cell is a large UO_2 grain surrounded by the ceramic wall, a cell in a metallic micro-cell pellet consists of many small conventional UO_2 grains covered by the metallic wall.

Gas atoms behavior in the UO_2 grains in a specific metallic micro-cell would be the same as in the grains of the conventional UO_2 pellet. However, even if the release pathways are formed at the grain boundaries, due to the presence of the wall, gas atoms would be unlikely to be released to pellet outside as long as the wall is intact as shown in Fig. 5(a).

As in the case of the ceramic micro-cell pellet, similar questions would arise as to the role of the wall on gas release—would gas atoms diffuse into the metallic wall from the surrounding UO_2 grains? And, if so, would they form gas bubbles within the wall and then interconnect each other to make release pathways? By the same arguments applied to the ceramic micro-cell pellet, gas atoms produced in a specific cell would be almost impossible to get out of it by diffusion if cell wall remains intact during irradiation.

Still the gas release would be possible if there are some cells whose walls blocking the release would be partly unavailable. Then, gas atoms would be released either by diffusion shown as the black arrows or by through release pathways represented as the red arrows in Fig. 5(b).

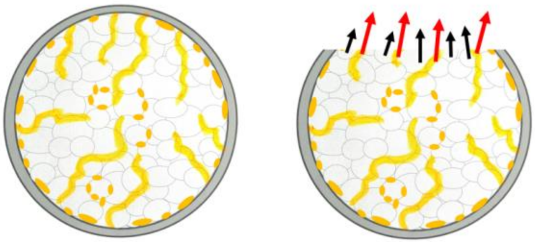

(a) (b)

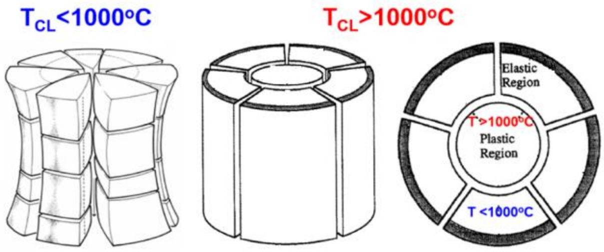
Fig. 5. (a) Small grains, gas bubbles at grain boundaries and release pathways that would likely to be observed in a metallic micro-cell pellet with intact wall during irradiation; (b) Gas release in a metallic micro-cell pellet in which cell wall does not exist in some region.

4. Estimation of Fission Gas Release in the Micro-cell Fuel Pellets

To estimate FGR in the micro-cell pellets, we need to know the fraction of the cells whose some areas are exposed to the free space. Two kinds of cells satisfy this requirement: the ones located at the top, bottom, and outer surfaces of the pellet during fabrication and the other ones located in the region through which radial cracks are created by tensile thermal stress upon starting reactor operation [9].

The number of cells located at the open surface during fabrication and the cells exposed to crack surfaces can be calculated on the basis of measurement for micro-cell pellet specimen [10]. Gas release could only take place in these cells because they are the only ones that are directly connected the open space. The number of cells exposed to the crack surfaces depends on how many radial cracks would be formed in the pellet during operation. According to a literature [9], the number of radial cracks in the pellet are about a half of linear power expressed in kW/m.

The cracks propagate to the region that is completely elastic, and it is known that UO_2 pellet exhibits elastic behavior for temperatures 1000-1200°C depending on the strain rate imposed on the pellet [11]. It is assumed that UO_2 is elastic at temperatures lower than 1000°C [12]. Fig. 6 shows the schematic geometry of a cracked UO_2 pellet on the basis of pellet centerline temperature. If centerline temperature is lower than 1000°C, all regions of the pellet would be cracked as in Fig. 6(a). For the centerline temperature higher than 1000°C, cracks would propagate only to the region whose temperature is 1000°C as indicated in Fig. 6(b), because plastic deformation occurs before ductile fracture in the central region [11].



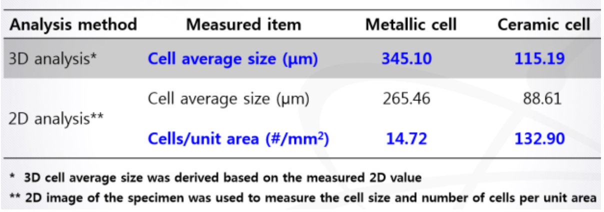
(a) (b)

Fig. 6. Illustration of the schematic geometry of a cracked UO_2 fuel pellet based on the centerline temperature of the pellet (T_{CL}): (a) $T_{CL} < 1000$°C; (b) $T_{CL} > 1000$°C.

Using the dimensions of micro-cell pellet, number of cracks, and centerline temperature, total surface area connected to the open space can be calculated. Then the number of cells in this total surface area would be obtained from the measurement which will provide the number of cells per unit area. Since the average size of the micro-cell in the specimen would be also measured, we can finally calculate the fraction of cells in a micro-cell pellet that would be available for gas release—a basis on which gas release can be estimated in the micro-cell pellets in comparison to the gas release in the UO_2 pellets.

Table 1 provides the average cell size and the number of cells per unit area that were measured for the two specimens: metallic micro-cell (5vol% Mo) and ceramic micro-cell (2vol% Si-Ti-O) [10]. Due to the small cell size of the ceramic micro-cell, the number of cells per unit area is about nine times higher than the metallic micro-cell. The average cell size was first measured by analyzing the specimen image two-dimensionally. Since this value does not represent the real cell size, it was converted into a three-dimensional one. The average cell size of the metallic and ceramic micro-cell is 345μm and 115μm, respectively.

Table 1. Average cell size and number of cells in the two specimens: metallic micro-cell (5vol% Mo) and ceramic micro-cell (2vol% Si-Ti-O) [10].



Analysis method	Measured item	Metallic cell	Ceramic cell
3D analysis*	**Cell average size (μm)**	**345.10**	**115.19**
2D analysis**	Cell average size (μm)	265.46	88.61
	Cells/unit area (#/mm²)	**14.72**	**132.90**

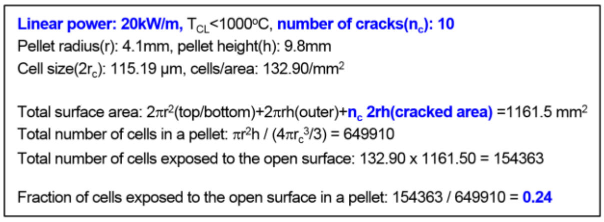
* 3D cell average size was derived based on the measured 2D value
** 2D image of the specimen was used to measure the cell size and number of cells per unit area

4.1 FGR in the Ceramic Micro-cell Pellet

Now we are in a position to estimate qualitatively how much fission gas would be released in a ceramic micro-cell pellet in comparison to a conventionally UO_2 pellet. First, using the measured values given in Table 1, the fraction of the cells connected to the open space is calculated by the procedure shown in Table 2.

The fraction of the exposed cells was calculated for the following conditions: linear power of the pellet is 20kW/m, which would produce ten radial cracks; centerline temperature is lower than 1000ºC so that the cracks can propagates to the centerline as shown in Fig. 6(a).

Table 2. Procedure for calculating the fraction of the cells exposed to the open surface in the ceramic micro-cell pellet of Table 1.



Linear power: 20kW/m, T_{CL}<1000ºC, **number of cracks(n_c): 10**
Pellet radius(r): 4.1mm, pellet height(h): 9.8mm
Cell size($2r_c$): 115.19 μm, cells/area: 132.90/mm^2

Total surface area: $2\pi r^2$(top/bottom)+$2\pi rh$(outer)+**n_c 2rh(cracked area)** =1161.5 mm^2
Total number of cells in a pellet: $\pi r^2 h$ / ($4\pi r_c^3/3$) = 649910
Total number of cells exposed to the open surface: 132.90 x 1161.50 = 154363

Fraction of cells exposed to the open surface in a pellet: 154363 / 649910 = **0.24**

Then what does it mean that the fraction of the cells exposed to the open surface in the ceramic micro-cell pellet is 24%? It is a general consensus that all grains in the conventional UO_2 pellet are connected to the open space once the release pathways are formed at their grain boundaries, implying the fraction of grains exposed to the open space is 100%. This means that all the fission gases produced in all the grains have a chance to be released. So if we denote the gas release fraction in the conventional pellet as f_{UO_2}, the maximum release rate possible in the ceramic micro-cell pellet would 0.24 f_{UO_2}. And probably the gas atoms in the remaining 76% cells would not likely to be released due to the presence of the cell walls.

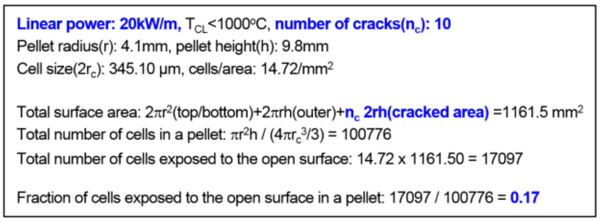
Even in the cells connected to the open space, as we see in Fig. 4(b), all the gas atoms in these cells would not have a chance to be released because the diffusion direction of the atoms is random. This would mean that only a part of the gas atoms in these cells, which would move towards the open space could be released, resulting in the gas release rate less than 0.24 f_{UO_2}.

4.2 FGR in the Metallic Micro-cell Pellet

As for the metallic micro-cell pellet, the fraction of cells that are connected to the open space is 17% as shown in Table 3.

By applying the same argument for the ceramic micro-cell pellet, we can say that the gas release rate in the metallic micro-cell pellet would be less than 0.17 f_{UO_2}. If we consider enhanced thermal conductivity of the metallic micro-cell pellet—50% higher than UO_2 pellet, the gas release rate would be further reduced for the condition of the same linear power.

Table 3. Procedure for calculating the fraction of the cells exposed to the open surface in the metallic micro-cell pellet of Table 1.



Linear power: 20kW/m, T_{CL}<1000ºC, **number of cracks(n_c): 10**
Pellet radius(r): 4.1mm, pellet height(h): 9.8mm
Cell size($2r_c$): 345.10 μm, cells/area: 14.72/mm^2

Total surface area: $2\pi r^2$(top/bottom)+$2\pi rh$(outer)+**n_c 2rh(cracked area)** =1161.5 mm^2
Total number of cells in a pellet: $\pi r^2 h$ / ($4\pi r_c^3/3$) = 100776
Total number of cells exposed to the open surface: 14.72 x 1161.50 = 17097

Fraction of cells exposed to the open surface in a pellet: 17097 / 100776 = **0.17**

5. Conclusion

Considering the microstructure of the micro-cell pellets and the gas release mechanism in the UO_2 pellet, gas release rates in the micro-cell pellets have been analyzed qualitatively. The analysis shows that gas release rates in the micro-cell pellets would be very low compared to UO_2 pellet for the following conditions: the cell walls would remain intact during irradiation, and the release pathways would not be formed in the walls. Then, for the same operating conditions, the release rates in the ceramic micro-cell and in the metallic micro-cell pellet would be less than 24% and 17%, respectively, of the UO_2 pellet's release rate.

Both in-pile and PIE data are required for understanding gas release mechanism and for the realistic modeling of gas release in the micro-cell pellets.

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