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Effect of the Size Distribution of Pu-rich Particles on Fission Gas Release in MOX Fuel

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Abstract

The effect of Pu inhomogeneity on fission gas release in MOX fuel has been analyzed by considering the size distribution of Pu-rich particles. Pu-rich particles are divided into a certain number of groups depending on their size. Then a gas release model, which uses the concept of the equivalent spherical cell composed of an equivalent spherical particle and the surrounding UO_2 matrix, is applied to calculate the gas release in each group of particles. The present analysis shows that the more Pu is accumulated in larger Pu-rich particles, the more fission gas is released for the same operating conditions. On the other hand, this effect decreases with burnup because at high burnup many gas atoms would be available for release at grain boundaries with the release paths already having been established. It is concluded that the present model explains why there exists some experimental evidence that fission gas release rates are enhanced in LWR MOX fuel compared with conventional UO_2 fuel under similar operation conditions.

1. Introduction

The MOX fuel for LWRs is fabricated either by direct mechanical blending of UO_2 and PuO_2 powders in proportions of the desired Pu content or by two stage mixing, where the master blend of intermediate Pu content prepared in the first step is mixed with the proper amount of pure UO_2 in the second stage to get the final Pu content. Therefore, it would be inevitable that incomplete mixing of Pu exists in MOX fuel, resulting in some number of Pu-rich particles with higher Pu content than the average for a fuel pellet. In addition, examination of the fabricated MOX pellet indicates that Pu-rich particles have a size distribution that is characteristic of its fabrication method. There is some experimental evidence that fission gas release rates are enhanced in LWR MOX fuel compared with conventional UO_2 fuel under similar operation conditions. The enhancement of gas release in MOX fuel may be attributed to incomplete Pu mixing along with slightly lower thermal conductivity and higher reactivity later in life. Since the last two factors can be considered in terms of fuel temperature, it is required to take into account the inhomogeneity effect of MOX fuel on fission gas release.

2. Modeling

2.1. Distribution and Division of Pu-rich Particles

It is assumed that the size distribution of Pu-rich particles $f(D_{agg})$ displayed in Fig.1(b), which is obtained from a sample of the specimen of Fig.1(a), can represent the one for a MOX fuel rod. It is also assumed for simplicity that the Pu-content in each Pu-rich particle is the same irrespective of its size and the Pu-content in the UO_2 matrix is uniform across the fuel pellet. In reality, the Pu content in the Pu-rich particles would also have some distribution with e_a being the average Pu content. Then from the conservation of Pu mass in the sample, we get

$$e_p V_{mea} = e_m \left(V_{mea} - V_{agg}\right) + e_a V_{agg}, \tag{1}$$

where e_p is the average Pu content in the sample, V_{mea} is the volume of the specimen used for measuring the size distribution of Pu-rich particles, e_m is the Pu content in the matrix, V_{agg} is the total volume of Pu-rich particles in the specimen, and e_a is the Pu content in the Pu-rich particles.

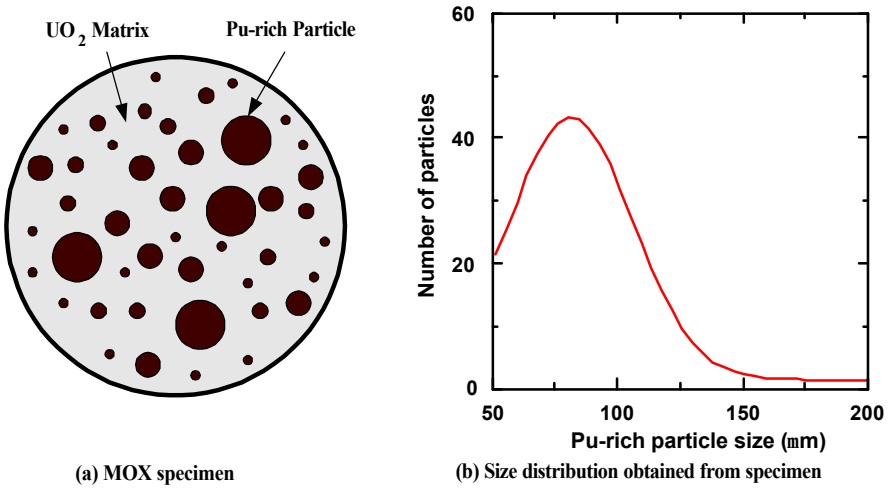


Fig.1. One example of the distribution of Pu-rich particles in MOX fuel.

Eq.(1) is then reduced to

$$V_{mea} = V_{agg}\left(\frac{e_a - e_m}{e_p - e_m}\right). \tag{2}$$

The Pu-rich particles are divided into g groups as shown in Fig.2 according to their sizes. Then the numbers of the Pu-rich particles corresponding to a particle diameter of D_{agg}^{1}, D_{agg}^{2} and D_{agg}^{g} are denoted as $f(D_{agg}^{1})$, $f(D_{agg}^{2})$, and $f(D_{agg}^{g})$, respectively. Here the sum of all Pu-rich particles should satisfy the following restraint:

$$N = \sum_{i=1}^{g} f(D_{agg}^{i}), \tag{3}$$

where N is the total number of Pu-rich particles retained in the sample specimen. If we represent V_{agg} using the particle diameter and number of particles corresponding to its diameter using the assumption that all Pu-rich particles are of a spherical shape,

$$V_{agg} = \sum_{i=1}^{g} \frac{4}{3} p \left(\frac{1}{2} D_{agg}^{i} \right)^3 f(D_{agg}^{i}) = \frac{p}{6} \sum_{i=1}^{g} \left(D_{agg}^{i} \right)^3 f(D_{agg}^{i}) . \quad (4)$$

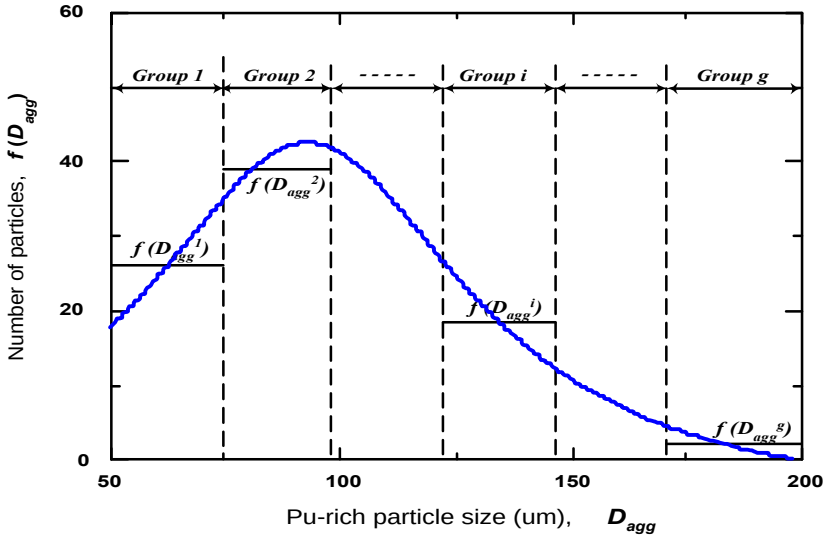


Fig.2. Division of Pu-rich particles into g groups according to their sizes.

2.2. Diameter of Equivalent Cell

The concept of the equivalent spherical cell, which is composed of an equivalent spherical particle with a diameter of $D_{eq}^{i} = D_{agg}^{i} + 2 \cdot L_{rec}$ and the surrounding UO_2 matrix [1], is used as shown in Fig.3. Here D_{eq}^{i} and D_{agg}^{i} represent the diameters of an equivalent spherical particle and a Pu-rich particle for the i-th group, respectively, and L_{rec} is the recoil length of fission products of about 6 μm. The diameter of an equivalent spherical cell D_{cell}^{i} for the i-th group is defined in such a way that the Pu mass in each equivalent cell is equal to the sum of the Pu mass in a Pu-rich particle and that in the UO_2 matrix. Then the diameter of an equivalent cell is derived using the assumption that the Pu contents in the Pu-rich particles are the same irrespective of the group to which they belong and those in the matrix regions are also the same for all groups:

$$D_{cell}^{i} = D_{agg}^{i} \left(\frac{e_a - e_m}{e_p - e_m} \right)^{1/3} . \quad (5)$$

If we denote the total volume of all the equivalent cells in the specimen as V_{cell}, then V_{cell} is

$$V_{cell} = \sum_{i=1}^{g} \frac{4}{3} p \left(\frac{1}{2} D_{cell}^{i} \right)^3 f(D_{agg}^{i}) = \frac{p}{6} \sum_{i=1}^{g} \left(D_{cell}^{i} \right)^3 f(D_{agg}^{i}) . \quad (6)$$

Then the combination of Eq.(6) with Eqs.(4) and (5) gives

$$V_{cell} = V_{agg}\left(\frac{e_a - e_m}{e_p - e_m}\right). \tag{7}$$

It is shown from Eqs.(2) and (7) that the volume of the specimen is equal to that of the total equivalent cells. This means that, if the concept of an equivalent spherical cell is introduced, the entire volume of the specimen is considered in the present analysis without leaving any of it untreated.

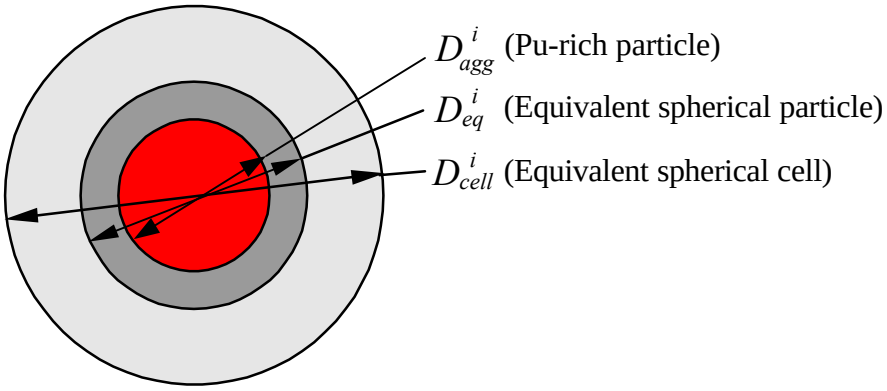


Fig.3. An equivalent spherical cell for a Pu-rich particle of the i-th group.

2.3. Fission Density in Each Region

The local fission density in the MOX fuel pellet, F_{loc}, is derived from the local power density using the assumption that an energy of 200 MeV is produced per fission. Then the fission densities in the equivalent particle, F_{agg}^{i}, and in the matrix, F_{m}^{i}, for the i-th group would be proportional to their Pu contents in each region, and hence they can be calculated as follows:

$$F_{agg}^{i} = \left(\frac{e_a}{e_p}\right) F_{loc}, \tag{8}$$

$$F_{m}^{i} = \left(\frac{e_m}{e_p}\right) F_{loc}. \tag{9}$$

The fission density in the equivalent particle, F_{eq}^{i}, for the i–th group is obtained in such a way that the total fission density in the equivalent particle is equal to the sum of the fission densities in the Pu-rich particle and that in the matrix retained between D_{eq}^{i} and D_{agg}^{i}. Then the following conservation is obtained:

$$F_{eq}^{i}\frac{4}{3}p\left(\frac{1}{2}D_{eq}^{i}\right)^3 = F_{agg}^{i}\frac{4}{3}p\left(\frac{1}{2}D_{agg}^{i}\right)^3 + F_{m}\frac{4}{3}p\left\{\left(\frac{1}{2}D_{eq}^{i}\right)^3 - \left(\frac{1}{2}D_{agg}^{i}\right)^3\right\}. \tag{10}$$

The combination of Eq.(10) and Eqs.(8) and (9) yields the fission density in the equivalent particle, $F_{eq}^{\ i}$, for the i-th group as follows:

$$F_{eq}^{\ i} = \left\{ \frac{e_m}{e_p} + \left(\frac{e_a}{e_p} - \frac{e_m}{e_p} \right) \frac{\left(D_{agg}^{\ i} \right)^3}{\left(D_{eq}^{\ i} \right)^3} \right\} F_{loc} . \tag{11}$$

2.4. Number of Equivalent Cells

If a MOX fuel rod is divided into a certain number of axial segments and a segment is also partitioned into a given number of radial rings with equal volume for performance analysis, the number of equivalent cells for i-th group, $n(D_{cell}^{i})$, contained in one radial ring is required to calculate the total gas release from that ring. The cell number for the i-th group, $n(D_{cell}^{i})$, is expressed by

$$n(D_{cell}^{\ i}) = f(D_{agg}^{\ i}) \frac{V_{ring}}{V_{mea}}, \tag{12}$$

where V_{ring} is the volume of one radial ring of an axial segment. If a fuel rod is divided into both M_a axial segments and N_r radial rings, V_{ring} is

$$V_{ring} = \frac{V_{rod}}{M_a N_r}, \tag{13}$$

where V_{rod} is the fuel rod volume. If Eqs.(2) and (13) are inserted into Eq.(12), $n(D_{cell}^{i})$ is expressed by

$$n(D_{cell}^{\ i}) = \frac{f(D_{agg}^{\ i})\, V_{rod}}{V_{agg}\, M_a N_r} \left(\frac{e_p - e_m}{e_a - e_m} \right), \tag{14}$$

where V_{agg}, which is the total volume of Pu-rich particles contained in V_{mea}, is obtained by

$$V_{agg} = \sum_{i=1}^{g} \frac{4p}{3} \left(\frac{1}{2} D_{agg}^{\ i} \right)^3 f(D_{agg}^{\ i}) = \frac{p}{6} \sum_{i=1}^{g} (D_{agg}^{\ i})^3 f(D_{agg}^{\ i}) . \tag{15}$$

Therefore, combining Eqs.(14) and (15), we obtain the number of equivalent cells for the i-th group $n(D_{cell}^{i})$ contained in one radial ring:

$$n(D_{cell}^{\ i}) = \frac{f(D_{agg}^{\ i})}{\left\{ \frac{p}{6} \sum_{i=1}^{g} (D_{agg}^{\ i})^3 f(D_{agg}^{\ i}) \right\}} \cdot \frac{V_{rod}}{M_a N_r} \cdot \left(\frac{e_p - e_m}{e_a - e_m} \right), \tag{16}$$

2.5. Gas Release in an Axial Segment

The amount of gas release, $REL_{cell}^{\ i}$, in an equivalent cell for the i–th group is calculated as follows:

$$REL_{cell}^{\ i} = REL_{eq,gr}^{\ i} \times \frac{V_{eq}^{\ i}}{V_{eq,gr}^{\ i}} + REL_{m,gr}^{\ i} \times \frac{V_{m}^{\ i}}{V_{m,gr}^{\ i}}, \qquad (17)$$

where

$REL_{eq,gr}^{\ i}$ = gas release from a grain in the equivalent particle contained in an equivalent cell for the i-th group,

$V_{eq}^{\ i} = \frac{4}{3}p\left(\frac{1}{2}D_{eq}^{\ i}\right)^3$: volume of the equivalent particle in an equivalent cell,

$V_{eq,gr}^{\ i} = \frac{4}{3}p\left(\frac{1}{2}D_{eq,gr}^{\ i}\right)^3$: volume of a grain in the equivalent particle of an equivalent cell,

$D_{eq,gr}^{\ i}$ = grain size in the equivalent particle of an equivalent cell,

$REL_{m,gr}^{\ i}$ = gas release from a grain in the matrix contained in an equivalent cell for the i-th group,

$V_{m}^{\ i} = \frac{4}{3}p\left\{\left(\frac{1}{2}D_{cell}^{\ i}\right)^3 - \left(\frac{1}{2}D_{eq}^{\ i}\right)^3\right\}$: volume of the matrix region in an equivalent cell,

$V_{m,gr}^{\ i} = \frac{4}{3}p\left(\frac{1}{2}D_{m,gr}^{\ i}\right)^3$: volume of a grain in the matrix of an equivalent cell,

$D_{m,gr}^{\ i}$ = grain size in the matrix of an equivalent cell.

Since fuel temperature in the equivalent particle would be higher than that in the matrix due to more fissions resulting from a higher Pu-content, it is very likely that $D_{eq,gr}^{\ i}$ would be larger than $D_{m,gr}^{\ i}$. But for simplicity, $D_{eq,gr}^{\ i}$ is assumed to be the same as $D_{m,gr}^{\ i}$. Then the total gas release in an axial segment, REL_{ax}, is calculated from

$$REL_{ax} = \sum_{i=1}^{g} n(D_{cell}^{\ i})\, REL_{cell}^{\ i}\ . \qquad (18)$$

The average gas release fraction, f, for a fuel pellet with g groups of Pu-rich particles is expressed as

$$f = \sum_{i=1}^{g} F_{agg}^{\ i} \cdot (REL_{eq}^{i} + REL_{m}^{i}) \Big/ \sum_{i=1}^{g} F_{agg}^{\ i} \cdot (PRO_{eq}^{i} + PRO_{m}^{i}), \qquad (19)$$

where F_{agg}^{i} is the fraction of Pu-rich particles corresponding to the i-th group of the total number of Pu-rich particles, REL_{eq}^{i} and PRO_{eq}^{i} are the released and produced amount of fission gas for an equivalent spherical particle of the i-th group, respectively, and REL_{m}^{i} and PRO_{m}^{i} are the corresponding values for the matrix region in an equivalent spherical cell.

2.6. Calculation Procedure

Based on the explanations described above, the calculation procedure for calculating fission gas release in MOX fuel considering the size distribution of Pu-rich particles caused by microstructure inhomogeneity is presented in Fig.4.

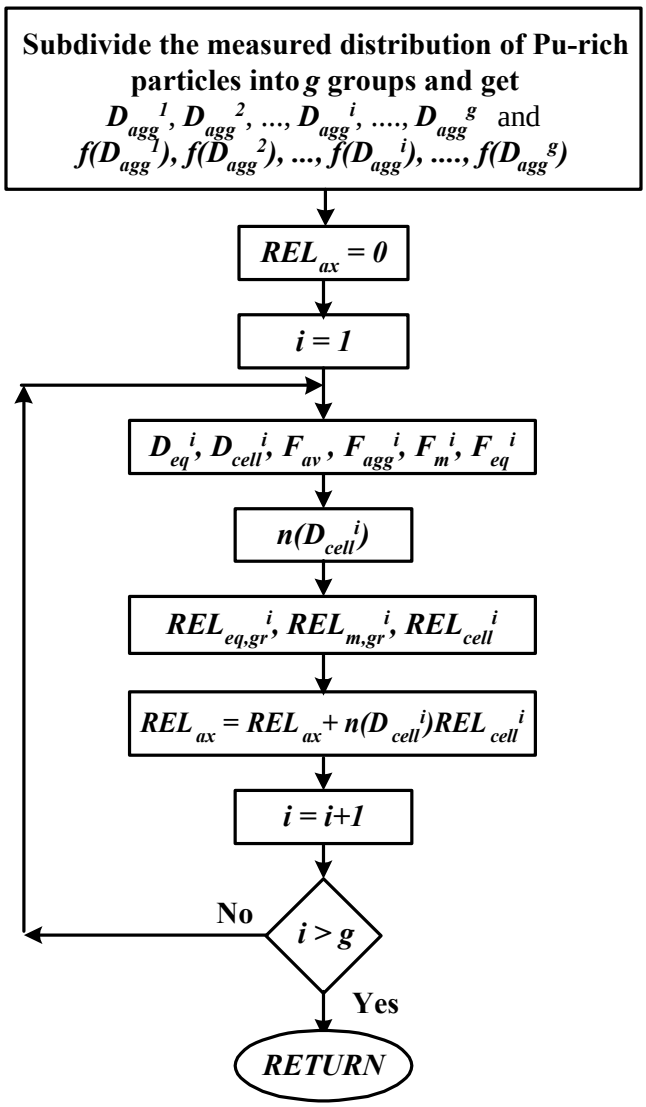
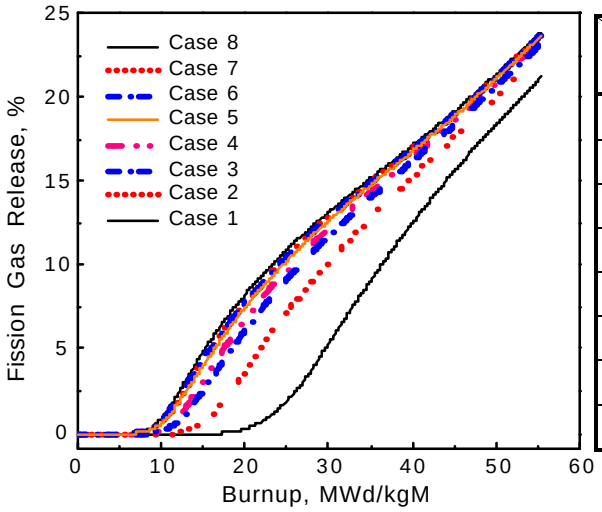


Fig.4. Flow chart for calculating fission gas release in one axial segment of a MOX fuel rod for g groups of Pu-rich particles.

3. Parametric Study

The model has been incorporated into a computer code COSMOS [2] and then a parametric study has been performed to investigate the inhomogeneity effect of MOX fuel on gas release for a constant fuel temperature of 900°C and a linear power of 250 W/cm. Grain sizes of both the Pu-rich particle and the UO_2 matrix are all assumed to be 15μm. In addition, the average Pu content of the fuel pellet is taken to be 0.06, while those for Pu-rich particles and UO_2 matrix are given as 0.23 and 0.04, respectively. Fig.5 shows the calculated results in ascending order of magnitude for the 8 cases given in the table below, where the combination of three sizes of Pu-rich particles (20, 30 and 40 μm) are considered. From these calculations, it is confirmed that the more Pu is accumulated in larger Pu-rich particles, the more fission gas is released for the same operating conditions. This is because more fission gas atoms would migrate to the grain boundaries in the larger Pu-rich particles than in the smaller ones due to a greater production of



	F^1_{agg} (20 μm)	F^2_{agg} (30 μm)	F^3_{agg} (40 μm)
Case 8	0.00	0.00	1.00
Case 7	0.00	0.50	0.50
Case 6	0.50	0.00	0.50
Case 5	0.33	0.33	0.33
Case 4	0.00	1.00	0.00
Case 3	0.50	0.50	0.00
Case 2	1.00	0.00	0.00
Case 1	UO_2		

Fig.5. Effect of the size distribution of Pu-rich particles on fission gas release.

gas atoms, thereby leading to the earlier formation of release paths in the grain boundaries and larger gas release in the Pu-rich particles. This phenomenon is dominant at low burnup region where the formation of release paths play a major role in determining the amount of gas released. On the other hand, the effect would decrease at high burnup fuel, because many gas atoms would be available for release at the grain boundaries with release paths already having been established. Consequently, the contribution of more gas production in the Pu-rich particles to gas release would be diminished with burnup. It is concluded from this parametric study that the present model explains why there exists some experimental evidence that fission gas release rates are enhanced in LWR MOX fuel compared with conventional UO_2 fuel under similar operation conditions.

4. Conclusion

A model has been developed that considers the effect of the size distribution of Pu-rich particles on fission gas release in MOX fuel. The model shows that the enhancement of gas release due the to inhomogeneity of MOX is dominant at low burnup where the formation of release paths plays a major role in determining the amount of gas released. On the other hand, the effect would decrease with burnup, because at high burnup many gas atoms would be available for release at the grain boundaries with the release paths already having been established.

Acknowledgements

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References

[1] Yang-Hyun KOO, Dong-Seong SOHN and P. STRIJOV, “A Fission Gas Release Model for MOX Fuel and Its Verification”, International Symposium on MOX Fuel Cycle Technologies for Medium and Long-Term Deployment, Vienna, Austria, May 17-21, 1999.

[2] Yang-Hyun KOO, Byung-Ho LEE and Dong-Seong SOHN, Journal of the Korean Nuclear Society, **30** (1998) 541-554.