Effect of Fission Product Bombardment on Structural Materials in Molten Salt Reactor Environments

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1. Introduction

It has been more than 60 years since the technology for molten salt reactors (MSRs) was first introduced. Initially MSRs demonstrated the possibility of applying thermal-spectrum and graphite-moderation concepts. In recent years, R&D has focused on the development of fast-spectrum MSR concepts, which combine the generic assets of fast neutron reactors with those associated with molten salts as a fuel and coolant. These MSRs can benefit from some advantages over solid-fueled systems, such as overcoming the difficulties of solid fuel fabrication, increasing the direct heat production in the mixed-fuel, reducing the risk of a loss of coolant, *etc.* Development of MSR technology is underway in Korea as well as other countries.

As a variety of MSR design concepts are considered depending on the situation of each country, we are developing MSRs that use a liquid fuel in the form of chloride salt and fast neutron spectrum. From the viewpoint of materials degradation, the primary concern is that the radioactive fission products (FPs) which are dissolved in the primary coolant system are always in contact with the containment vessel. That can cause corrosion and related problems. According to the Molten Salt Reactor Experiment performed in 1960s at Oak Ridge National Laboratories, it was found that the grain boundary embrittlement of nickel-based alloys was associated with the FP tellurium [1-3]. Also, the displacement damage to the vessel material due to fast neutron irradiation is inevitable. In this work, potential damage to materials owing to energetic FPs was investigated by using simulation methods, which include the penetration of FPs and sputtering by energetic particles. Sputtering is the erosion of a material by particle bombardment, which results in the removal of surface atoms of a sample. To this purpose, we calculated the sputtering yield by energetic FPs and their range using a computer code.

2. Methods

When an incident ion penetrates a solid, it loses energy by a series of collisions with the atoms and electrons and comes to rest in the solid. The actual penetrated distance traveled by the ion is called the range, R. During the course of ion-solid interactions, lattice atoms can be ejected from the outer surface layers, which corresponds

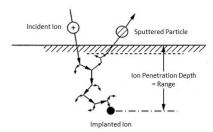


Fig. 1. Schematic of ion-solid interactions and the sputtering process

to sputtering. Fig. 1 shows a two-dimensional schematic view of an ion-solid interaction, as well as the sputtering process. Seen from Fig. 1, we can imagine that an energetic FP acts as an incident ion in MSR environment.

To evaluate the sputtering of a target and the range of FPs, a SRIM computer code was applied [4]. The SRIM calculates the sputtering yield, which is defined as the average number of sputtered target atoms per incident ion. In this work, the target material is SS304, which was assumed to consist of three elements of Fe, Cr, and Ni. In order to setup the input data for the SRIM, the information on the FPs for ²³⁵U fission is to be examined.

2.1 Fission Products from ²³⁵U fission

As a result of ²³⁵U fission with thermal neutrons, a fissioning nucleus should split approximately in half. However, fission is always asymmetric, so the elements and masses of the two products are different. The fission product yield from a ²³⁵U fission with a given atomic number is given in Fig. 2. The average total kinetic energy of two FPs depends on a neutron energy, which was set to be 168.5 MeV in this work. This value is appropriate for a thermal ²³⁵U fission reaction.

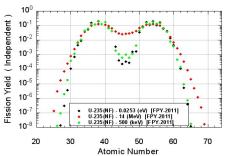


Fig. 2. ²³⁵U neutron-induced fission yields – JAEA Nuclear Data Center [5]

Seen from Fig. 2, the atomic numbers of lighter FPs are primarily between 34 and 41 and those of heavier ones are between 50 and 56. Five fission reactions with relatively higher yield probability were selected among a number of ²³⁵U-fission reactions, which are listed in Table I with the kinetic energy for each FP. Besides, other reason for the selection is that the FPs of Te and Xe are known to be detrimental to the degradation of structural materials. The values of FP kinetic energy are used as input to the SRIM code.

Table I: ²³⁵U thermal fission reactions and FPs kinetic energies

²³⁵ U fission reactions	FP Kinetic Energy (MeV)			
⁹⁰ Sr + ¹⁴⁴ Xe + 2n	Sr	103.7	Xe	64.8
⁸⁷ Br + ¹⁴⁶ La + 3n	Br	105.6	La	62.9
⁹⁶ Rb + ¹³⁷ Cs + 3n	Rb	99.1	Cs	69.4
⁹⁷ Zr + ¹³⁷ Te + 2n	Zr	98.7	Te	69.8
⁹² Kr + ¹⁴¹ Ba + 2n	Kr	104.3	Ba	64.2

3. Results

3.1 Fission Product (Ion) Range

In MSR environments, the ion range, R plays a role in understanding radiation damage to structural materials. It is important to predict the migration of ions penetrated into the reactor materials. The R depends on the energy, mass, atomic number of ions as well as on those of the target material. We calculated the R for the fission products tabulated above with an incident angle of 0 degree – perpendicular to the surface. As an example, the R of two FPs for Te and Zr is plotted in Fig. 3. The calculated R for the given fission products is summarized in Table II.

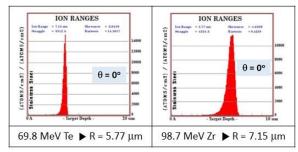


Fig. 3. Ranges of fission products of Te/Zr (incident angle = 0°)

Table II: Calculated ranges of fission products

FP	Sr	Br	Rb	Zr	Kr
R (μm)	6.98	7.46	7.12	7.15	7.12
FP	Xe	La	Cs	Te	Ba
R (μm)	4.89	5.41	5.70	5.77	5.48

3.2 Sputtering Yield

It is likely that sputtering by energetic FPs can lead to negative effects on the vessel wall, affecting material ejection and surface morphological changes. Highenergy FPs can create surface roughness, pits and craters, which are dependent on the energy and incident angle of the particles. For the given conditions in Table I, the sputtering yield, Y was evaluated by varying the incident angles. Examples of the Y estimation for Te and Zr are shown in Fig. 4, where Y increases with the incident angle of FPs. Sputtering yields for FPs incident to the surface of SS304 with the incident angle of 45°, which were calculated from the SRIM, are given in Table III. Relatively heavier elements with lower kinetic energy tend to cause higher sputtering of surface atoms.

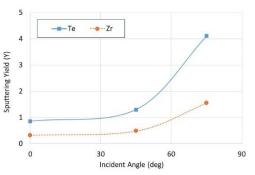


Fig. 4. Incident angle dependency of sputtering yield, Y by FPs (Te and Zr)

Table III: Sputtering yield by FPs at the incident angle of 45°

	Sputtering Yield, Y						
Sr	0.442	Xe	1.575				
Br	0.349	La	1.679				
Rb	0.398	Cs	1.411				
Zr	0.492	Te	1.306				
Kr	0.385	Ba	1.613				

4. Discussion

It is probable that radiation damage to structural materials would occur in the MSR environments when mixed molten salts are used as fuel and coolant together. The energetic FPs are always in contact with the vessel, which can lead to the penetration of FPs into the vessel materials and the sputtering of surface atoms. In this work, we evaluated the range of FPs and their sputtering yield using the SRIM computer code. Although the FP ranges are not so long, the diffusion of FPs inside materials plays a role in microstructural changes. The sputtering by FPs can cause the ejection of surface atoms and changes in surface morphology. Such a latent damage due to FPs might affect the integrity of MSR structures over a long period of operation, not in the short-term. In order to evaluate the radiation damage to materials caused by energetic FPs in a quantitatively, we need to know the neutron spectra for MSR, which is not available, currently being designed. As a preliminary study, the potential effect of FP bombardment on materials was investigated in this study.

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