Cesium Diffusion Tests in Graphite with Optimized ICP-MS Measurement Conditions

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1. Introduction

Graphite has been used widely in the nuclear industry since Chicago pile-1, which was the first nuclear reactor in the world, because graphite has excellent properties as a moderator. High temperature gas-cooled reactors (HTGR) also use graphite as a moderator. The fuel design of HTGR is based on the concept of tristructural-isotropic (TRISO) particles buried into a graphite matrix. The graphite matrix works as not only the moderator but also a structural material for the TRISO particles. The coating layers of TRISO particles act as barriers of fission products that are produced from the fuel kernel. When the coating layers fail to prevent the release of the fission products due to cracking, fission products might be adsorbed in the graphite matrix. Some fission products such as cesium (Cs), silver (Ag), and strontium (Sr) are likely to cause contamination since they are released out to primary coolant channel. Therefore, evaluating the behavior of the fission products in the graphite matrix is important for safety concerns.

In the authors' previous work [1-2], the diffusion test samples preparation, the preliminary diffusion tests, and the results analysis methodology have been studied. In these studies, ICP-MS measurements were conducted to analyze the samples quantitatively. But the results showed highly scattered data even if the samples are similar. To improve the reliability of the ICP-MS measurements results, optimization of the measurement has been conducted.

2. Optimizing the ICP-MS measurement conditions

2.1 Background

ICP-MS measurements have been conducted to analyze the diffusion test samples quantitatively. The previous results showed a cesium (Cs) range of 146-652 ppm in the graphite matrix, the data is highly scattered. The solid samples have to be converted to liquid because ICP-MS measurements are applied on liquid samples only. To convert the samples, they were ground as powder. The powders were treated to dissolve out Cs from the graphite to the solution, which is called the 'pre-treatment' step. It was shown that the conditions of the pre-treatment caused the data scatter as expected. In the authors' previous work [2], the powder size effect was studied mentioning that the effect is not only from the powder size, but from other parameters that are studied in this paper.

2.2 ICP-MS optimization

In addition to the powder size effect, the powder mass effect, the pre-treatment temperature, and time were studied. Only Cs can be dissolved into the solution, and the graphite remains. Therefore, when the powder size is larger, the diffusion distance is longer and the dissolution rate is decreased. In Table 1, the powders which are coarser than 212 μm showed a lower concentration of Cs. In contrast, finer powders, which have a size range of 63~106 μm, showed a much lower concentration.

Table 1. ICP-MS results with variation of powder size range

Powder mass (mg)	Particle size (μm)	Concentration (ppm)
68.6	>212	175.1
33.2	106~212	418.8
1.2	63~106	85.4
53.4	<63	424.1

The powder mass was a vague parameter as the additional tests were conducted. The target masses

were 1 mg, 10 mg, and 100 mg, the powders were prepared to investigate the effect of mass on the Cs concentration. As shown in Table 2, the powders that have 10 mg and 100 mg masses showed similar concentrations. But the results of the 1 mg powders showed much lower concentrations. It means that both the weighting error of powder mass and the low level of Cs quantities affect the results of ICP-MS measurements.

Table 2. ICP-MS results with variation of weight of powders

Test set	Powder mass (mg)	Concentration (ppm)
1	1	78.6
	11.2	405.9
	101	394.9
2	0.8	198.8
	9.8	303.8
	93	292.7

The pre-treatment temperatures were 50°C, 100°C, 150°C, and 200°C and the results of ICP-MS measurements are shown in Table 3. The concentration results are proportional to the pre-treatment temperature. This means that the temperature affects the activation of Cs and accelerates the dissolution process.

Table 3. ICP-MS results with variation of pre-treatment temperature

Pre-treatment temperature (°C)	Concentration (ppm)
50	162.5
100	186.9
150	239.1
200	308.3

The results of ICP-MS at 200°C with different pre-treatment time are listed in Table 4. When the pre-treatment time is 30 minutes, the Cs concentration is lower than all the other cases, which have relatively similar concentrations.

Table 4. ICP-MS results with variation of pre-treatment time at 200°C

Test set	Holding time (hours)	Concentration (ppm)
1	0.5	291.5
	4	463.8
	12	466.0
2	1	495.9
	2	505.5
	3	513.3
	4	516.3

3. Conclusion

In this paper, the results of optimized ICP-MS measurements, which were used to analyze the diffusion test samples quantitatively, were obtained and discussed. To conduct ICP-MS measurements, the samples have to be converted to liquid in the pre-treatment process. The target of optimization was to change the pre-treatment conditions such as powder size, powder mass, pre-treatment temperature, and pre-treatment time to optimum values. The ICP-MS measurements will be conducted following the optimized conditions based on the results of this study. The results show that optimum conditions to get the intended results include using powders finer than 212 μm and larger than 10 mg and holding the pre-treatment time to be longer than 1 hour at 200°C.

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