Reaction Characteristics of $CsNO_3$ by Different Adsorbents

Dasom Yang*, Jinmyeong Shin, and Sangho Na

Korea Atomic Energy Research Institute, 111, Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, Republic of Korea

*dsyang@kaeri.re.kr

1. Introduction

Semi-Volatile cesium(Cs) can be released during the operation of radioactive waste incineration and melting of radioactive metal waste at nuclear facilities. Cs is the most significant semi-volatile fission product which is highly radioactive and heat generating fission product. Cs is very leachable under disposal environment. ^{134}Cs, ^{135}Cs and ^{137}Cs are radioactive isotopes, ^{135}Cs has a long half-life of about 2.3×10^6 years, while ^{134}Cs has a short half-life of about 2years. ^{137}Cs has an intermediate half-life of 30.2years. Among these isotopes, ^{137}Cs is a major concern because of its high abundance, intense gamma radiation and high heat generation.

Cesium oxide can be readily incorporated into aluminosilicate compounds by thermal reaction, and the resulting cesium aluminosilicate compounds are stable. This stability of cesium in aluminosilicate structures may be exploited in the fixation of nuclear waste, by the trapping radioactive cesium into stable cesium aluminosilicate phases.

This study was carried out to analyze the reaction characteristics of $CsNO_3$ with sintered kaolin, fly ash and pyrophyllite, respectively.

2. Experimental

Kaolin, fly ash and pyrophyllite were sintered at 1,150℃ for 2 hours, cooled and powdered for the experiment. The analyzed particle size of kaolin, fly ash, pyrophyllite used in this study is around 0.15 mm (200 mesh), 0.2~0.35 mm (70 mesh), 0.15 mm (200 mesh), respectively. The particle size of $CsNO_3$ (Sigma Aldrich) is 0.2 ~0.35 mm (70 mesh). The reaction experiments of $CsNO_3$ with sintered kaolin, fly ash and pyrophyllite were maintained at 1,000℃ for 3 hours under air condition. Melting point of $CsNO_3$ is 414℃ and it begins to volatilize near at 700℃. And 98% of $CsNO_3$ is volatilized.

The $CsNO_3$ and kaolin, fly ash and pyrophyllite were mixed with Cs, Al and Si in the mole ratio of 1:1.02:1.07, 1:1.02:1.76, 1:1:3.60, respectively.

The trapping quantity of cesium on adsorbents and weight change were calculated as follows.

Table 1. Reaction quantity of cesium on different adsorbents

Adsorbent	Before reaction(g)	After reaction(g)	Reaction quantity (g-Cs/g-adsorbent)
Kaolin	1.5593	1.2511	0.78
Fly Ash	1.5272	1.2678	0.45
Pyrophyllite	1.5390	1.3427	0.34

The weight loss of before and after reaction of kaolin is 19.5%, fly ash is 17% and pyrophyllite is 12.8%. Based on these results, the reaction quantity of kaolin, fly ash and pyrophyllite is 0.78 g-Cs/g-kaolin, 0.45 g-Cs/g-fly ash, 0.34 g-Cs/g-pyrophyllite, respectively. As shown in Table 1, reaction quantity of cesium indicated the formation of cesium aluminosilicate compounds of three adsorbents exposed to cesium vapors.

In this study, to confirm the presence of cesium reacted with adsorbent, cesium peak is analyzed by XRD (X-Ray Diffraction Analyzer). The XRD was applied to analyze the phases of the reaction product. The scanning rate was 0.2°/min and the angle, 2θ, was within the range of 10° to 90°.

The cesium reaction product of sintered kaolin gave XRD peaks that indicate Cs-nepheline ($CsAlSiO_4$) and pollucite ($CsAlSi2O_6$) phases as shown in Fig. 1. As shown in Fig. 1, XRD pattern confirmed that Cs-nepheline ($CsAlSiO_4$) phase was mainly formed.

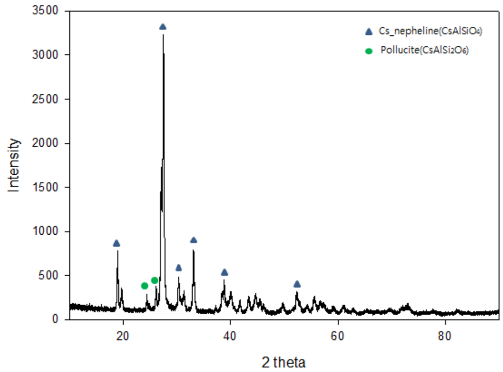

Fig. 1. X-ray diffraction patterns of cesium reaction product of sintered kaolin.

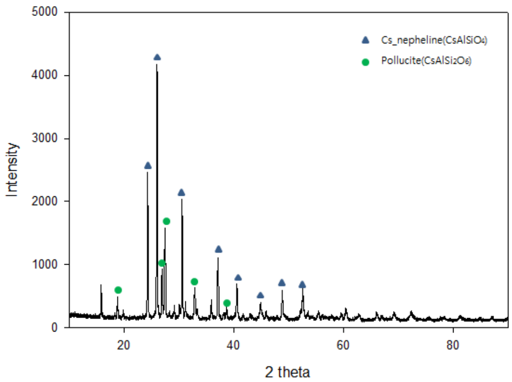

Fig. 2. X-ray diffraction patterns of cesium reaction product of sintered fly ash.

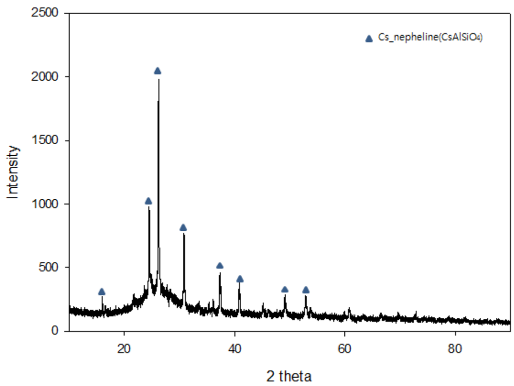

Fig. 3. X-ray diffraction patterns of cesium reaction product of sintered pyrophyllite.

Fig. 2 shows the XRD pattern of the cesium reaction product of sintered fly ash. The XRD pattern confirmed pollucite phase and Cs-nepheline phase were formed.

Fig. 3 shows the X-ray diffraction patterns of cesium reaction product of sintered pyrophyllite. The XRD pattern confirmed the existence and the crystal structure of the single phase Cs-nepheline.

The SEM image results are shown in Fig. 4. SEM image indicates that sintered kaolin has platy particles stuck to together to form agglomerates and irregular shapes (Fig. 4(a)). SEM image also indicates that sintered kaolin reacted with $CsNO_3$ is bulky crystals with a rough surface (Fig. 4(b)).

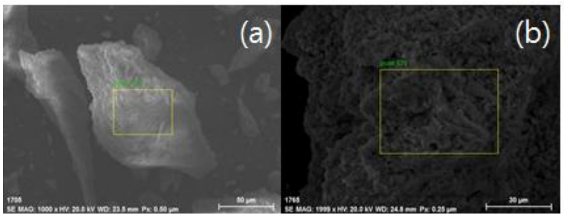

Fig. 4. (a) SEM image of sintered kaolin, (b) SEM image of sintered kaolin after reaction with cesium.

3. Conclusion

Reaction characteristics of $CsNO_3$ with sintered kaolin, fly ash and pyrophyllite were studied respectively. The high trapping quantity of these sintered adsorbents is believed to due to the formation of pollucite ($CsAlSi_2O_6$) phase and Cs-nephline ($CsAlSiO_6$) of the reaction products. Because of their high performance, easy availability and low cost, kaolin, fly ash and pyrophyllite are expected to be utilized as adsorbents for trapping gaseous cesium.

REFERENCES

[1] J.J. Park, J.M. Shin, M.S. Yang, K.S. Chun, H.S. Park, “Trapping characteristics for gaseous cesium generated from different cesium compounds by fly ash filter”, IAEA-SM-357/71.