Development of Off-Gas Capturing System from Pyroprocessing

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1. Introduction

An accumulation of spent fuel has brought a considerable interest due to its energy and environmental issue. Pyroprocessing has been investigated in Korea Atomic Energy Research Institute (KAERI) which reduces the accumulated spent fuel and increases the efficiency of fuel cycle. In head-end process of pyroprocessing, spent fuels are thermally treated to prepare suitable UO_2 pellets for electrochemical separation of TRU(transuranic elements), and various radioactive gases are released from spent fuel at high temperature. Within these gases, Cs-137 and I-129 are semi-volatile gases which are highly radioactive and hazardous to environment, and therefore it is imperative to develop efficient method for capturing cesium and iodine. In this study, a kg-scale off-gas capturing system was developed to adsorb cesium and iodine, and the adsorption characteristics were investigated.

2. Methods and Results

2.1 Trapping Filters

It is well known that cesium highly reacts with aluminosilicate to form $CsAlSi_2O_6$ or $CsAlSiO_4$, and these structures are very stable in storing for long period. KAERI had developed a silica-alumina(SA) filters and had confirmed that it is effective in adsorbing cesium [1]. It is advantageous to fabricate SA filters because the size, shape, and porosity can be easily controlled by using suitable support material. In the case of iodine adsorption, iodine highly reacts with silver to form AgI which is a stable structure. KAERI had developed iodine adsorption process using silver-functionalized zeolite(AgX) as an adsorbent. The SA filters were prepared from kaolinite ($Al_2Si_2O_5(OH)_4$) solution mixed with 2 wt% PVA (Fig. 1). The prepared solution was dispersed to cylindrical polyurethane sponge and for several times and then calcined at 1200 ºC for overnight in air. The fabricated SA filters were placed in horizontal alumina tube and reduced at 1000 ºC for overnight using 4%-H_2 based Ar gas. In addition, AgX containing about 35 wt% were pretreated at 400 ºC in reducing condition prior to the adsorption test.

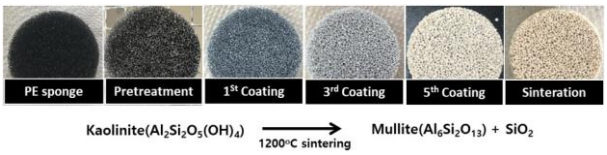

Fig. 1. Fabrication of SA filters from kaolinite

2.2 A kg-scale off-gas capturing system

The adsorption process was consisted of two different regions; vaporization and adsorption zones. Cs_2CO_3 and CsI were used as precursor for cesium and iodine, and they were placed in the alumina boat in the center of the vaporization zone and temperature was fixed to 1100 ºC. In the adsorption zone, two adsorbing columns were filled with SA and AgX adsorbents, and they were operated at 1000 ºC and 200 ºC, respectively. The uncaptured particles consisted of cesium and iodine might pass through SA and AgX filters, and they were separated using ULPA(ultra-low particulated air) filter as a membrane. Water scrubbers were installed at the end of the adsorption process to trap remaining particles in the vent gas. The overall kg-scale off-gas capturing system were shown in Fig. 2.

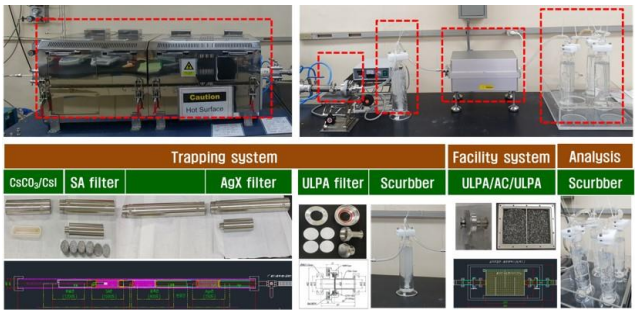

Fig. 2. A kg-scale off-gas capturing system

2.3 Characteristics of filters

The vaporized cesium reacts with SA filter by forming $CsAlSiO_4$ (Fig. 3), and the adsorption performance of ~0.99 g of Cs_2O per g of filter was obtained. In addition, the vaporized iodine was found to react with silver from AgX filter, leading AgI formation (Fig. 4).

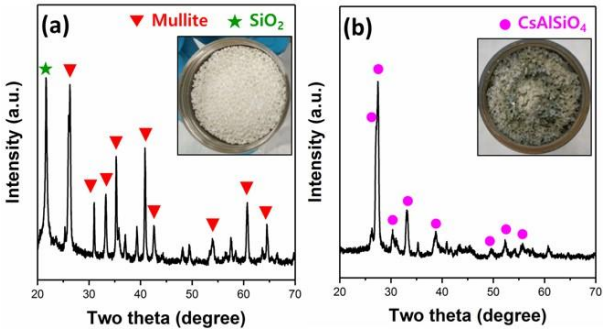

Fig. 3. XRD analysis for (a) pristine SA filter and (b) cesium reacted SA filter.

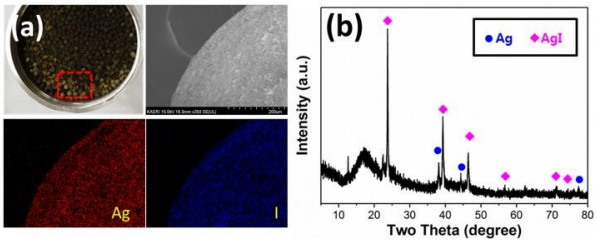

Fig. 4. (a) EDS mapping and (b) XRD pattern for AgX filter after I capture.

Moreover, cesium and iodine were found in ULPA filters confirmed from EDS mapping analysis, and it is attributed that uncaptured particulate matters from SA and AgX filters were efficiently trapped on ULPA filter.

2.4 Trapping efficiency

The trapping efficiency for the off-gas capturing system was calculated by comparing vaporized amount estimated from surrogate (Cs_2CO_3/CsI) and emitted amount analyzed from ICP-MS.

Table I: Trapping efficiency for cesium and iodine from liquid scrubber

	Amount Vaporized (g)	Amount emitted (g)	Trapping efficiency (%)
Cs	18.876	<0.03	>99.999999
I	1.683	<0.3	>99.9999

3. Conclusions

A kg-scale off-gas capturing system with SA filter and AgX was designed to capture cesium and iodine vaporized from head-end process for treatment of 5 kg of spent fuel and found to exhibit the trapping efficiencies of >99.999999% for cesium and >99.9999% for iodine respectively through verification tests.

REFERENCES

[1] Jae Hwan Yang et al., A kaolinite-based filter to capture gaseous cesium compounds in off-gas released during the pyroprocessing head-end process, Annals of Nuclear Energy, Vol. 103, p. 29, 2017.