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 CsAlSiO$_4$ or CsAlSiO$_6$, 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$_2$Si$_2$O$_5$(OH)$_4$) solution mixed with 2 wt% PVA (Fig. 1). The prepared solution was dispersed to cylindrical polyurethane sponge and then calcined at 1200 °C for overnight in air. The fabricated SA filters were placed in horizontal alumina tube and reduced at 1000 °C for overnight using 4%-H$_2$ based Ar gas. In addition, AgX containing about 35 wt% were pretreated at 400 °C in reducing condition prior to the adsorption test.

2.2 A kg-scale off-gas capturing system

The adsorption process was consisted of two different regions; vaporization and adsorption zones. Cs$_2$CO$_3$ 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 °C. In the adsorption zone, two adsorbing columns were filled with SA and AgX adsorbents, and they were operated at 1000 °C and 200 °C, 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.

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$_2$O 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).
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$_2$CO$_3$/CsI) and emitted amount analyzed from ICP-MS.

<table>
<thead>
<tr>
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<th>Vaporized (g)</th>
<th>Emitted (g)</th>
<th>Trapping efficiency (%)</th>
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</thead>
<tbody>
<tr>
<td>Cs</td>
<td>18.876</td>
<td>&lt;0.03</td>
<td>&gt;99.999999</td>
</tr>
<tr>
<td>I</td>
<td>1.683</td>
<td>&lt;0.3</td>
<td>&gt;99.9999</td>
</tr>
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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