Removal of Radionuclides from Molten LiCl-KCl Part 1: Cs, Sr and Ce removal by using a tablet-shaped inorganic material

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1. Introduction

LiCl-KCl waste generated from an electro-refining process for EBR-II spent fuel contains a series of radionuclides, different from a pyroprocess in Korea. As an immobilization method suggested by ANL, zeolite-4A is mixed with molten salt waste to produce saltoccluded zeolite that can be consolidated to a ceramic waste form (CWF, Glass-bonded sodalite) at about 915 °C by adding borosilicate glass; the mixing ratio of salt/zeolite/glass is about 1/9/3.33 on a weight basis [1]. Although the CWF is a practical example for the final disposal of the salt wastes, this approach seems to be an inefficient method in that the final waste volume is increased by about 10 times over that of the initial waste salt. For this reason, a selective removal of some fission product radionuclides from the molten salt phase is indispensable. The selective removal studies by using zeolite-4A were first carried out for the LiCl-KCl waste. Selective precipitations by phosphates or hydroxides were reported [2, 3]. In Korea, other technologies are under investigation for selective removal or recovery of clean LiCl-KCl salt.

Although II or III group in salt waste would be removed by precipitation, I group elements such Rb and Cs is not precipitated by phosphate, carbonate or oxide because of solubility in molten salt. Our research group intended to find new separation technologies for reducing final waste volume.

As a chemical approach, this study was focus on the development an effective separating agent. In order to find or develop a new separating agent, a material to selectively remove Cs is most necessary. In screening commercial zeolites or synthetic zeolites, we found that analcime (ANA) is very effective material for Cs removal from LiCl-KCl molten salt. This study investigated the removal ability of ANA and tried to fabricate a tablet-typed separating agent for the removal of radionuclides.

2. Methods and Results

Analcime was synthesized by hydrothermal reaction at 200°C. The synthetic procedure was referred from IZA(International Zeolite Association, <u>www.iza-online.org</u>). A removal experiment in the molten salts was performed in a batch type reactor. Concentrations of CsCl, SrCl₂ and CeCl₃ in the salts were about $0.1\sim1.0$ wt%. When a temperature of the salts reached 450°C, a fixed amount of separating agent was injected into the salts and then the salts was stirred at about 150 rpm for 120min. A small amount of molten salts $(0.1 \sim 0.3g)$ as a sample taken at a time step by using an alumina tube. The sample was dissolved in purified water and 0.1M HNO₃ solution, and the concentrations of Cs, Sr and Ce in this solution was analyzed by an ICP-AES and AAS. After a removal experiment of radionuclides, the use inorganic materials were identified by XRD analysis.

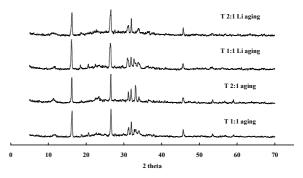


Fig. 1 XRD pattern of inorganic materials before heat treatme -nt

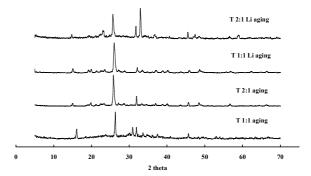


Fig. 2 XRD pattern of inorganic materials after heat treatment at 600°C

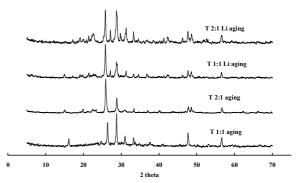


Fig. 3 XRD pattern of inorganic materials after molten LiCl – KCl phase at 450°C batch-type reactor

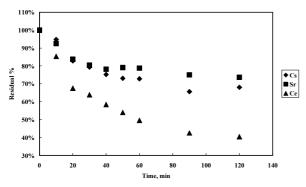


Fig. 4. Removal behavior of tablet-typed separating agent for each element(200g salt/5g SA, 1wt% CsCl, SrCl₂, CeCl₃)

Fig. 1,2 and 3 shows the XRD patterns of prepared separating agent before and after batch experiments. Fig. 4 indicated the removal behavior of tablet-typed separating agent for each element. The removal efficiency of Cs, Sr and Ce were about 35%, 25% and 60%, respectively.

3. Conclusion

As a chemical approach to remove radionuclides from molten salt, this study investigated the fabrication condition for tablet-typed separating agent and its removal behavior. The prepared material has an ability to capture I/II/III elements from molten salt.

Acknowledgments

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