

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

Mi Sook Son, *Hwan-Seo Park, In-Tae Kim, So Lim Kang

Korea Atomic Energy Research Institute, 1045, Daedeok-Daero, Yuseong-gu, Daejeon, 305-353, Korea

*Corresponding author: hspark72@kaeri.re.kr

1. Introduction

In the pyroprocess to recover uranium and transuranic elements, waste salts containing fission products are generated. The one is LiCl waste from an electro-reduction process and the other one is LiCl-KCl waste from an electro-refining process. These wastes are difficult to directly immobilize into durable matrix such as borosilicate glass or ceramic wasteform for final disposal due to their physico-chemical properties[1]. As a solution for this, the ANL(Argonne National Laboratory) suggested a method using a unique zeolite structure that can capture metal chlorides into its large cage; metal chloride-loaded zeolite-4A converts into sodalite at about 915°C. Although this wasteform shows high leach-resistance, the waste volume greatly increases[2]. For this reason, it is necessary to find a method to selectively remove fission products or recover non-radioactive LiCl or LiCl-KCl, lowering final waste volume. The INL(Idaho National Laboratory) suggested a selective removal by using zeolite-4A for LiCl-KCl waste. Also, a precipitation by using metal phosphates in melt or hydroxide in aqueous solution was tried by other researchers. In KAERI, oxide precipitation by sparging O₂ gas or melt crystallization are under investigation. This study is focus on the development of new separating material (SA) to remove radionuclides from LiCl waste. As a commercial material, zeolite-4A suggested at INL cannot be applied to LiCl waste because its structure is decomposed into a useless compound for a selective removal[3]. In this study, the removal behavior of radionuclides at 650°C was investigated by using a new tablet-typed SA.

2. Methods and Results

2.1 Method

SA for a removal radionuclides in molten salt chlorides(LiCl; m.p. 600°C) was made as follows. First, reagents such as TEOS(tetraethyl orthosilicate, Aldrich, Germany), AlCl₃·6H₂O, HNO₃, ethanol and purified water were mixed in a ratio. A hydrogel was formed as a result of the mixing and it dried at 110°C in an electric oven for 2 days. The dried material was rubbed up with ethanol and purified water and then was deformed to a tablet-shaped. A tablet type of an inorganic material used in this study was treated to remove hydrated water at 600°C for 2 hrs.

A removal experiment of CsCl, SrCl₂ and CeCl₃ in the molten salts was performed in a batch type reactor. Concentrations of CsCl, SrCl₂ and CeCl₃ in the salts were about 0.1~1.0wt%. When a temperature of the salts reached 650°C, a fixed amount of SAs were injected into the salts and then the salts was stirred at about 150 rpm for 120min. A small amount of molten salts(0.1~0.3g) 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 used SAs were identified by XRD analysis.

2.2 Fabrication of tablet-typed separating agent



Fig. 1 Photograph of SA before(a) and after (b) heat treatment at 600°C.

Commercial zeolites frequently used in a selective removal from aqueous wastes are decomposed into Li-aluminosilicates when contacting a molten LiCl; zeolite-4A structure was not changed in molten LiCl-KCl. For this reason, our research group intended to develop a new separating agent(SA) that is chemically active to radionuclides in molten LiCl.

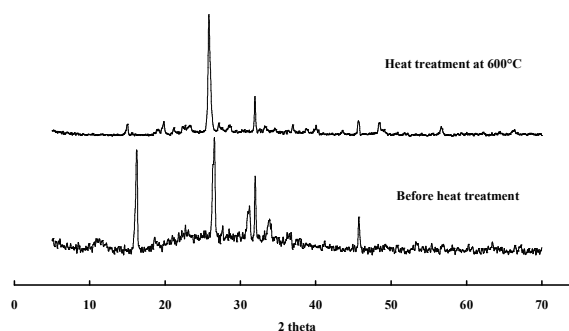


Fig. 2 XRD pattern of SA before(a) and after (b) heat treatment at 600°C.

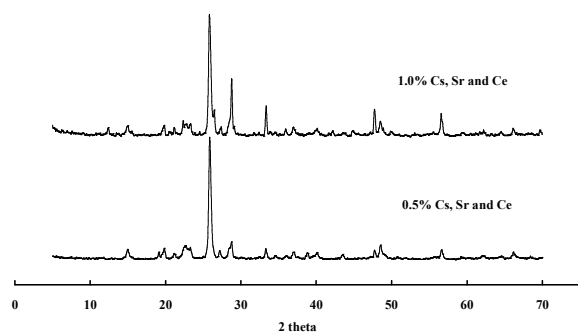


Fig. 3 XRD pattern of SA obtained from a batch experiment for a removal test of Cs, Sr and Ce in the molten LiCl at 650°C.

Fig. 1 and 2 show the prepared SAs and their XRD patterns before and after heat-treatment. The XRD patterns shown in Fig. 2 indicated the small contents of seed crystal/additive and it means that amorphous parts in SA take the removal ability.

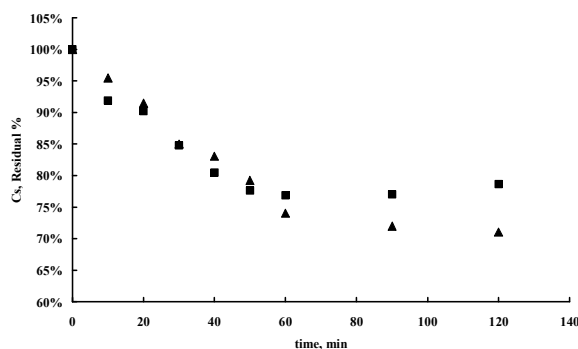


Fig. 4 Removal results of Cs from the molten LiCl phase at 650°C by using SA(200g salt/5g SA, ▲: 0.5%, ■: 1.0% initial wt% of CsCl in LiCl salt).

Fig. 3 shows the XRD patterns of SA after a batch experiment and Cs removal behavior with time. Under given conditions, 25~35% of Cs was removed in 2h. The removal experiments for the other radionuclides are going on.

3. Conclusion

Our research group intended to develop a new separating agent applicable to molten LiCl waste. This study fabricated a tablet-typed SA and investigated the removal behavior for radionuclides. More experiments are necessary to understand the chemical behavior between SA and LiCl wastes.

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