Effect of Initial Nuclide Concentration on the Purification of LiCl Waste Salt by Layer-Melt Crystallization Process

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1. Introduction

The wastes salt generated from pyrochemical process after sufficient use of chloride salts as molten state electrolytes are LiCl waste salt from the electrolytic reduction process, and LiCl-KCl eutectic waste salt from the electrorefining and electrowinning processes. In particular, the waste salts have highly heat generative group I & II fission products such as Cs, Sr and Ba nuclides and highly radioactive generative rare earth fission products. Therefore, the fission products within the waste salt should be separated and concentrated in small volume to reduce the volume of final waste [1].

In this study, the effect of initial concentration on separation performance of group I & II nuclides from LiCl waste was investigated using layer-melt crystallization at various initial concentrations of nuclides.

2. Experimental

Layer-melt crystallization process was carried out in a 3 Kg/batch lab-scale apparatus as shown in Fig. 1(a). The apparatus consisted of crystallization furnace, melting furnace (recover purified LiCl from the crystallizer plate), crystallizer module and Ar circulation system (as coolant), data acquisition system. All the parts are located in a glove box maintained Ar atmosphere to prevent moisture and oxygen. LiCl crystals were grown by layer-melt crystallization, where crystallization plate is rectangular shape with thickness of 15 mm, 60 mm wide, and 200 mm height. Module of crystallizer consisted of three thermocouples, three crystallization plates [2]. The temperature monitoring system is applied to recover high purity LiCl crystal by stopping the crystallization process when the crystal is grown properly without over-growing. The stop point of process was determined by temperature monitoring of around crystallizer module: surface of crystallization plate (Crystallizer TC), center of between crystallization plate (Center TC) and molten salt (Salt TC), as shown in Fig. 1(b).

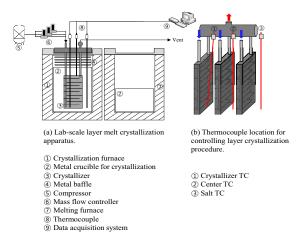


Fig. 1. Scheme of lab scale layer-melt crystallization apparatus.

The initial concentration ratio of cesium and strontium/barium nuclides are set based on the properties of LiCl waste salt generated after oxide reduction process. These tests were performed under various initial concentrations. After crystallization process, concentration of impurities in purified LiCl salt was analyzed using an Inductively Coupled Plasma Spectrometer.

3. Results and discussion

Solid-liquid phase diagram was identified separation possibility and temperature condition of crystallization process. Fig. 2 shows the solid-liquid phase diagram for the LiCl-CsCl-SrCl₂-BaCl₂ system drawn using the FactSage software. As shown in the Fig. 2, there is temperature range that exists only in a solid state of LiCl at 500~600°C. The solidus temperature of LiCl is decreased as the initial concentration of cesium and strontium/barium nuclides in the LiCl increased. The effect of the

initial concentration of impurity nuclides on the separation efficiency can be seen in Fig. 3. The results show that the separation performance decreased when the initial concentration of cesium and strontium/barium nuclides in the LiCl increased. This is because at a high initial concentration, many impurities of mother liquid are entrapped in the crystal layer under the same growth conditions.

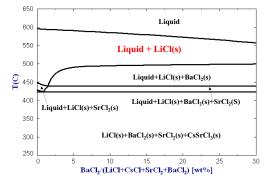


Fig. 2. Phase diagram of LiCl-CsCl-SrCl₂-BaCl₂ system.

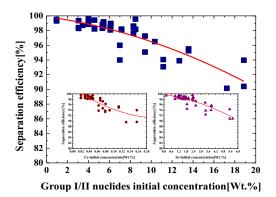


Fig. 3. Effect of the initial nuclides concentration on the separation efficiency.

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