

Application of Zeolite on Removal of Radionuclides (I/II) within Waste LiCl Salt

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1. Introduction

The pyrochemical treatment process of a spent nuclear fuel, which is mainly composed of an electrolytic reduction of an oxide fuel to a metallic form, an electrorefining of the reduced metal ingot and an electrowinning, has been developed in Korea Atomic Energy Research Institute (KAERI) since 1997. In the electrolytic reduction process, the spent oxide fuel is reduced into uranium metal in a molten LiCl salt. During the reduction process, the molten LiCl salt, containing chloride forms of radioactive fission products such as alkali metal, alkaline-earth, and rare-earth, is generated as a waste salt. This waste salt is water-soluble and readily radiolyzed to yield interstitial halogens and metal colloids. Thus, KAERI has focused on a minimization of a waste salt generation, such as a recycling after a removal of radioactive nuclide (I/II) using zeolite.

This project is based on two ideas, first, zeolite has an ion-exchange capability even at a high temperature, and second, zeolite is also used for a final waste form of this kind waste salt. Fission products, especially high heat-generation elements such as cesium (Cs) or strontium (Sr), which remain mainly(→)mainly remain in the waste LiCl salt, have been known to be removed by an ion-exchange between those nuclide ions and Li-ion (or Na-ion) on the lattice and alpha cages of zeolite. Up to now, this kind application has conceptually suggested, but not been successfully applied to Li-based system that runs at 650°C.

2. Experimental

2.1 Batch Ion-Exchange

The experiment was performed with a batch ion-exchange apparatus maintained at 650°C under an argon atmosphere. Before an experiment, zeolite 4A was dried at 550°C for 20 h. The samples were analyzed by Inductively Coupled Plasma – Atomic Emission Spectroscopy (ICP-AES) for Cs, Sr and Ba, Atomic Absorption Spectroscopy (IS) for Cl. The mixing ratios of LiCl to zeolite were 4.5 to a maximum of 25. The initial concentrations were minimum values, which were calculated by the OREGON Code after one batch reduction, to maximum values after 160 batch operation.

2.2 Column Ion-Exchange

Column ion-exchange experiment was performed in a vessel maintained at 650°C. Argon gas was used for purging and an injection. The molten salt was prepared in a 400-ml tank, and injected into 1-inch diameter and 4-inch height zeolite column. The facial velocity of the molten salt was from 1.24 to 4.0 cm/min.

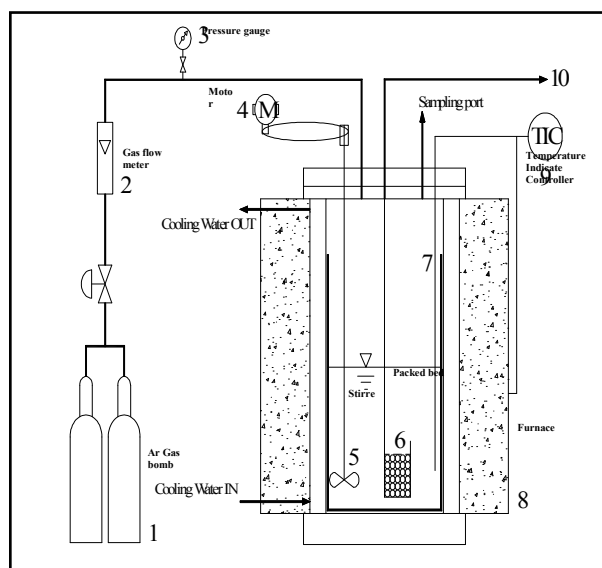


Fig. 1. Schematic diagram of batch ion-exchange experiment.

3. Results and Discussion

3.1 Batch Ion-Exchange

Fig. 2 shows the experimental result for Cs containing LiCl salt. X-axis means the operation time and the Y-axis means the Cs concentration in the molten LiCl salt. If the removal by an ion-exchange happened, the Cs concentration decreases gradually. However, Cs concentration increased or did not change, instead of a decrease. The initial Cs concentration of the lowest result (represent as dot in Fig. 2) was 280 ppm Cs in the molten salt, which is supposed to be a Cs concentration after just one batch reduction. In spite of such a low Cs concentration, the Cs concentration in the molten LiCl salt did not changes(→)change, meaning no removal by a zeolite ion-exchange at 650°C.

3.2 Column Ion-Exchange

The column ion-exchange experiment result showed that the Cs concentration in the molten LiCl salt was not

changed even at a very low Cs initial concentration (the result figures are not showed here).

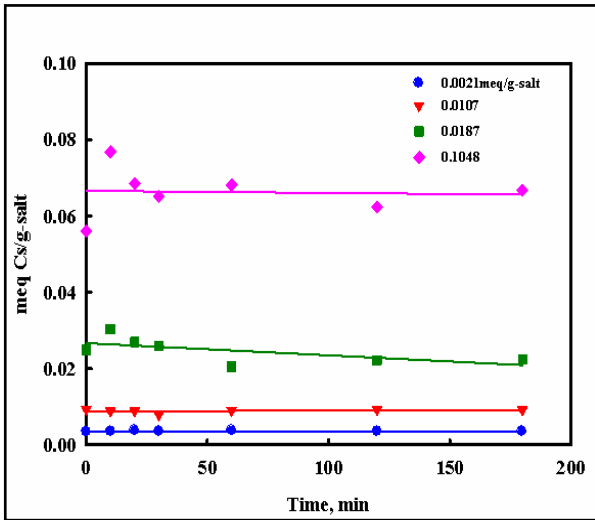


Fig. 2. Cs concentration change during the batch ion-exchange experiment

3. Conclusions

A salt-zeolite exchange was applied to remove the radioactive heat-generation elements such as Cs, Sr, and Ba in a molten waste LiCl salt at 650°C. The experimental result showed that the radionuclide (I/II) was hardly removed by using zeolite A since zeolite lost its ion-exchange capability at this temperature

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