Radionuclides Removal Using Cancrinite and Chalcogel Sorbents

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1. Introduction

Oxidation layer of primary system of nuclear power plant consists of radionuclides and metals like Fe, Ni, Cr etc. The layer is generally removed by chemical oxidation-reduction decontamination (CORD) process which is a kind of chemical decontamination method. Then, the remaining contaminants (radionuclides and metals) in waste solution is treated by passing ion exchange resin to remove the metal ions. However, the spent resin is not easy to dispose because of the presence of some organic materials and large volume of the spent resin which is about 4500 L. Therefore, it is necessary to reduce the volume of the waste resin and develop the inorganic sorbents which can be easily used for waste form and disposal in repository.

In this study, we synthesized cancrinites and chalcogel to remove the radionuclide and metal ions in simulated CORD waste solution condition. Cancrinites were expected to have high ion exchange capacity for heavy metal cations and anions due to its low Si/Al ratio and the presence of exchangeable anions in cage structure [1]. In addition, chalcogel was investigated to have high specific surface areas and multifunctional characteristics for use of sorbent. Our main objective is to synthesize inorganic sorbents (cancrinites and chalcogel) for removal of radioactive wastes in CORD waste solution and understand the removal mechanism.

2. Experimental section

2.1 Synthesis of cancrinites

Cancrinites were synthesized using hydrothermal synthesis method at low temperatures of 200℃ and autogenous pressure in 125 mL Teflon-coated steel Parr reactor. For carbonate cancrinite, 4 g of kaolinite($Al_2O_7Si_2 \cdot 2H_2O$) and 0.5 M of sodium carbonate were mixed in 90 mL of 8 M of sodium hydroxide solution. The synthesis was performed for 45 h inside an oven. All the final products were centrifuged by 3000 rpm for 10 min, washed to decrease pH to around 7 using deionized (DI) water and dried for 24 h at 80℃. The final white powder products were analyzed by X-ray diffraction for mineral identification. In case of nitrate cancrinite, 1 M of sodium nitrate and 72 h heating was used instead of sodium carbonate and 45 h heating process for carbonate cancrinite synthesis.

2.2 Synthesis of chalcogel

Chalcogel was synthesized using Sol-Gel method. For synthesis of Co-Pb-MoS chalcogel, 0.14 mmol of $Co(NO_3)_2 \cdot 6H_2O$ and 0.06 mmol of $Pb(CH_3COO)_2 \cdot 3H_2O$ were dissolved in 4 mL of formamide. Ammonium tetrathiomolybdate (0.2 mmol) was dissolved in 2 mL of formamide. Then, Co-Pb precursor solution was added into the tetrathiomolybdate solution without stirring or heating for 7 days. Then, mixed solution was

decanted and the chalcogel was washed by DI water for 5 days to remove the residual ions with changing the DI water every 24 h. Then, the chalcogel was divided to two parts of particles. One was dried using Freeze dryer (0.1 mBar, -50℃), while another was dried at room temperature without oven.

3. Results

The XRD results of synthesized cancrinites are shown in Fig. 1 and 2. The XRD patterns of synthesized cancrinites (nitrate and carbonate) well match with the references. Synthesized chalcogel shows black color as shown in Fig. 3. After the freeze drying process, the chalcogel becomes powder which makes the surface area increased. Additional analysis (SEM-EDS, BET, etc.) is also planned for chalcogel characterization.

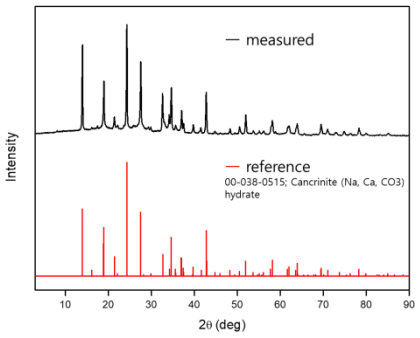

Fig. 1. XRD pattern of the carbonate cancrinite with reference.

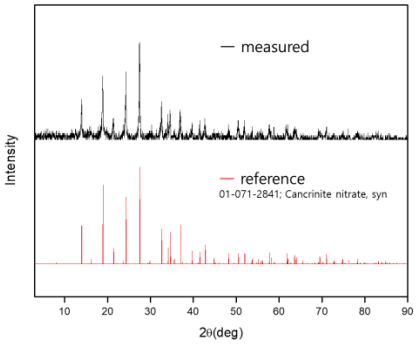

Fig. 2. XRD pattern of the nitrate cancrinite with reference.

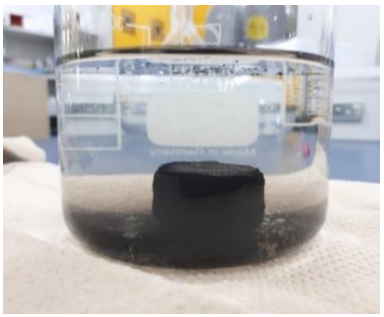

Fig. 3. Solvent exchanging process of the chalcogel.

4. Conclusions

The adsorption using the synthesized cancrinites and chalcogel would be conducted under the aqueous condition prepared to mimic the CORD waste solution. In carbonate cancrinite, because of carbonate ion playing a role for a pH buffer, the carbonate cancrinite was still stable and could be used even at low pH conditions (pH=1-3), while nitrate cancrinite was dissolved in low pH condition. For the chalcogel, ability to stand at low pH should be tested. In addition, both of the inorganic sorbents would be investigated for removal of radionuclides.

ACKNOWLEDGEMENT

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REFERENCES

[1] Hackbarth, K. et al., “Synthesis and crystal structure of carbonate cancrinite $Na_8[AlSiO_4]_6CO_3(H2O)_{3.4}$, grown under low-temperature hydrothermal conditions.”, Microporous and Mesoporous materials, 1999. 30(2-3): p.347-358.