Removal of Iodide Using Sn-Zeolite Under Aqueous Condition

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1. Introduction

In the world, electric power production of nuclear power plants (NPP) has been progressed for the past decades. Moreover, electric power production from NPP for continuous energy can be beneficial without producing green-house gas (CO₂). However, all radioactive wastes generated from NPP operation should be securely and safely managed during the nuclear fuel cycle. Especially, the waste includes radioisotope such as ¹²⁹I and ¹³¹I whose half-lives are $t_{1/2} = 1.57 \times 10^7$ years and $t_{1/2} = 8.02$ days, respectively[1]. These iodine has harmful effects to humans through external and internal exposure which can bring burns to the skin and affect the thyroid gland. Therefore iodine needs to be removed safely.

In this study, we used the HY zeolite and Sn ion for capturing iodide from solution waste. HY Zeolite is eco-friendly materials with having high sorption capacity, thermostability, ion exchange, and high pore size as porous material. And Sn ion has chemical interaction with iodide ion and economically affordable. Therefore, the objective of this study is to investigate the capturing radioactive iodine from solution using surface modified HY zeolite with Sn.

2. Materials and Methods

2.1 Synthesis of HY zeolite

HY zeolite was prepared from a commercial NH_4 zeolite (powder, Sigma-Aldrich Co. LLC) through heating method. NH_4 zeolite was calcined at 500 °C in furnace under a dry air atmosphere for 8 h. And then, HY zeolite was formed after vaporization of NH_3 gas[2].

2.2 Surface modification of HY zeolite

Sn-HY zeolite was formed using the surface modification method. First, Tin(II) chloride (powder, 99.99% SnCl₂, Sigma-Aldrich Co. LLC) was dissolved in Deionized water (DIW) to prepare 0.03 M solution. Synthesized HY zeolite was mixed with SnCl₂ solution in solid to liquid ratio of 1g:100mL at room temperature under stirring vigorously for 8 h. And then, the resultant suspension was filtered by vacuum filter. Finally, Sn-HY zeolite was dried at 110 °C in oven for 8 h[2]. Sn-13X and Sn-NaY zeolite were also formed by the surface modification method to compare with Sn-HY zeolite.

2.3 Iodine Capturing Test

 ^{127}I Nonradioactive iodine, (as surrogate for radioactive iodine), solutions were prepared in crimp-top glass amber 10mL vials (Wheaton) for iodine batch experiments. 10mL Iodide solution (¹²⁷I concentration = 4ppm and pH = 3) was reacted with adsorbent (50mg). The pH was adjusted by 5% HNO3 solution. The samples were covered by the lids and placed in a basic shaker (os-3000) for continuous mixing at 130rpm for 10 days, as kinetic experiment which is to conclude equilibration time and reaction rate of iodine. After experiments, the aqueous samples were collected at different reaction times and analyzed with inductively coupled plasma mass spectroscopy (ICP-MS, NexION 300D) to measure iodine concentrations. The capture efficiency of adsorbent for each iodine species was determined by Eq (1).

$$Efficiency\,(\%) = \left[\left(C_i - C_s\right)/C_i\right] \times 100\tag{1}$$

where C_i is initial iodine concentration (ppm) which was determined with blank sample and C_s is iodine concentration (ppm) in each sample at different reaction times after filtration[3].

3. Results and Discussion

The result of iodide capturing test is shown in Figure 1. The iodide adsorption capacity on Sn-HY zeolite is 64.12%. It is higher than Sn-13X (15.76%) and Sn-NaY (52.55%) zeolite. It is demonstrated that Sn is well coordinated with HY zeolite for capturing iodide, compared with 13X and NaY zeolite.

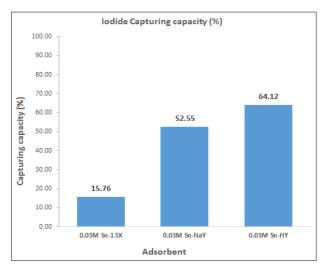


Fig. 1. Iodide capturing capacity using 0.03 M Sn-13X, Sn-NaY, and Sn-HY.

4. Conclusion

Iodide is captured by synthesized Sn-HY zeolite better than Sn-13X and Sn-NaY zeolite. Additional study is required to investigate the mechanism between iodide ion and Sn-HY zeolite whether it is chemical sorption or physical sorption. In addition, although iodide removal efficiency of Sn-HY zeolite is good in this study, the efficiency of Ag-zeolite is little bit higher than Sn-HY. So, it is still needed to upgrade performance in capacity and other properties like economic feasibility.

5. Reference

 Yujiro Watanabe, Toshiyuki Ikoma, Hirohisa Yamada, Yasushi Suetsugu, Yu Komatsu, Geoffrey W. Stevens, Yusuke Moriyoshi, and Junzo Tanaka, "Novel Long-Term Immobilization Method for Radioactive Iodine-129 Using a Zeolite/Apatite Composite Sintered Body", ACS applied materials & interfaces, 1 (7), 1579-1584 (2009).

- [2] Yi-Hsiu Chen, Shing-Jong Huang, and Ben-Zu "Reversal Wan, of Zeta Potential of after Proton-Type Zeolite Y Surface Modifications in Sodium Ion Aqueous Solutions", Insudtrial and Engineering Chemistry Research, 55, 1921-1928 (2016).
- [3] Brian J. Riley, David A. Pierce, Jaehun Chun, Josef Matyáš, William C. Lepry, Troy G. Garn, Jack D. Law, and Mercouri G. Kanatzidis, "Polyacrylonitrile-Chalcogel Hybrid Sorbents for Radioiodine Capture", Environmental Science and Technology, 48 (10), 5832-5839 (2014).