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Performance Enhancement of Adsorbent for Capturing Radioactive Technetium

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1. Introduction

The technology designed to recycle accumulated spent nuclear fuel is pyroprocessing. However, various fission products (FPs) are generated during the process. Among these FPs, technetium-99 (Tc-99) is a gaseous nuclear isotope with a long half-life and high mobility, and it should be captured strictly to prevent radioactive contamination of the environment.

Calcium oxide (CaO) is an excellent adsorbent to remove Tc-99 generated from used nuclear fuel. Previous studies identified optimal temperatures for Re (surrogate for Tc) capture at either 400 ℃ or 900 ℃, with a significant decrease in adsorption performance observed at 500–800 ℃. To broaden the range of adsorption temperatures, we evaluated the capturing performance by adjusting the amount of isopropyl alcohol (IPA), a pore-forming agent added during the synthesis of CaO adsorbents.

Experimental tests were conducted at various temperatures to determine the optimal IPA content for the most effective capture performance.

2. Methods and Results

2.1. Preparation of CaO adsorbent

First, 15 g of calcium acetate ($Ca(CH_3COO)_2$) is added to 45 g of distilled water, and the mixture is stirred until fully dissolved using a magnetic stirrer. Then, 13 g of ethylene glycol ($(CH_2OH)_2$) is added, and the mixture is stirred at 50 ℃ for 10 minutes. Subsequently, 80 g of calcium hydroxide ($Ca(OH)_2$) is added, and the mixture is stirred until a paste with the desired viscosity is formed. Finally, IPA (isopropyl alcohol), serving as the pore-forming agent, is added to the adsorbent paste, and the mixture is stirred until the desired viscosity is achieved once again. Six different adsorbent samples were prepared by varying the amount of the pore-forming agent. The adsorbents were named CP-0 to CP-125 based on the ratio of IPA to CaO paste.

2.2. Re adsorption experiment

The system was designed to examine the capture performance of CaO adsorbent for Re at different temperature. Re capture experiments were conducted based on the experimental conditions from previous research [1]. Using a mass flow controller (MFC), air was supplied to a quartz tube at a flow rate of 24 ml/min (linear velocity ~0.5 cm/s).

The experimental procedure is outlined as follows: the section containing the adsorbent was heated to 800 ℃ at a rate of 10 ℃/min and maintained for 1 h to perform pretreatment, removing impurities such as moisture and carbon CO_2. When the target temperature of the adsorbent was <800 ℃, it was cooled and maintained at that temperature. In case of the target temperature was >800 ℃, pretreatment heating was carried out at the target temperature. After 1 hh of pretreatment, the temperature of the section containing the reagents was then heated to 450 ℃ at a rate of 5 ℃/min. This temperature was maintained for 2.5 h to ensure the volatilization of all Re reagents. Following the completion of the experiment, air continued to flow during the natural cooling process.

2.3. Characteristics of the Re adsorbed CaO adsorbent

The result, presented in Figure 1, illustrate the Re adsorption performance of CaO adsorbents adsorbents based on the adsorbent type (CP-0 to CP-125) and capture temperature (400 to 900 ℃). When comparing the Re adsorption performance of the CP-50 adsorbent, prepared by adding 100 mL of IPA as used in previous studies, it is evident that the adsorption performance of the adsorbents with higher IPA content has improved in the temperature range of 400 to 700 ℃. Specifically, the highest adsorption performance is observed in the CP-100 sample at 500 ℃.

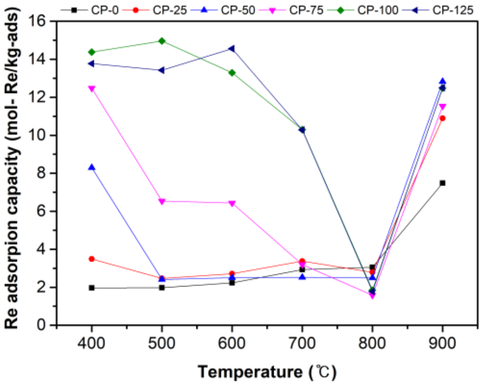

Fig. 1. Re adsorption performance of CaO adsorbent as a function of IPA/CaO paste ratio and capture temperature

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3. Conclusions

In previous studies, the optimal temperature for Re capture was found to be either 400 ℃ or 900 ℃, with a significant decrease in adsorption performance observed at between 500 ℃ and 800 ℃. To extend the range of adsorption temperatures, we evaluated the capture performance by adjusting the amount of pore-forming agent (IPA) during the synthesis of the CaO adsorbent. To determine the optimal IPA content for the best capture performance, Re adsorption experiments were conducted at different temperatures. The experimental results of this study suggest that the expanded temperature range could facilitate the separation of gaseous Tc, providing valuable insights for designing the capture process.

REFERENCES

[1] S.-M. Hong et al. "Development of a CaO-based pellet for capturing gaseous technetium-99 from spent nuclear fuel" Journal of Environmental Chemical Engineering (2022).