Evaluation of Trapping Characteristics for Volatile Kr/Xe Off-Gas

Seok-Min Hong, Jae Hwan Yang, and Yung-Zun Cho

Korea Atomic Energy Research Institute, Daedeok-daero989ben-gil 111,Yuseong-gu, Daejeon, Republic of Korea

seokminhong@kaeri.re.kr

1. Introduction

An accumulation of spent fuel has brought a considerable interest due to its energy and environmental issue [1]. Pyroprocessing has been investigated in Korea Atomic Energy Research Institute (KAERI) which reduces the accumulated spent fuel and increases the efficiency of fuel cycle. In head-end process of pyroprocessing, UO_2 pellets are produced from a fuel bundle and various radioactive gases from a spent fuel are released during thermal treatment. Within these gases, Kr is a volatile gas which has a long half-life of 11 years and high radioactivity, and therefore it is imperative to capture Kr [2]. Several methods such as cryogenic distillation, absorption using solvents, and adsorption on solids have been widely applied to Kr capture. Of these approaches, adsorption has many advantages such as its low energy-consumption processes and easy regeneration. In addition, Xe also can be released from the spent fuel and it inhibits Kr trapping performance due to its relatively high adsorption affinity. Therefore, Xe should be trapped even it has lower half-life and radioactivity compared to Kr. In this study, Kr/Xe adsorption process was designed and tested using a packed bed column filled with adsorbents. The Kr/Xe adsorption performances were determined from breakthrough curves obtained from a gas chromatography, and the effects of adsorption conditions on capacities were investigated.

2. Experimental Section

2.1 Preparation of Adsorption Test

The adsorbents were pretreated at 130 ^{o}C in a vacuum oven for overnight to remove moisture and impurities. The dried adsorbents were packed in a column of a diameter of 30 mm with flowing inert gas to prevent adsorbing water from atmosphere. The flow was 126 cm^3 min^{-1} and the pressure was 1 atm.

2.2 Gas Analysis

The gas chromatography (GC, DS Science, Republic of Korea) equipped with a pulsed discharge detector (PDD) was used to measure a gas concentration which passed through the adsorption column. A 30 m of GC column (ValcoPLOT Molesieve 5A) was installed. He gas was used as a carrier gas in GC, and Kr and Xe was calibrated using standard gases. The temperature of GC was maintained at 60 ^{o}C.

2.3 Breakthrough Curve

An adsorption capacity was calculated using a breakthrough time as follows:

$$q_b = \frac{F}{M}\int_0^{t_b}(Co - C)\mathrm{dt} \tag{1}$$

where q represents adsorption capacity (mmol kg^{-1}), F is a flow rate (cm^3 min^{-1}), M is a mass of adsorbent (g), t is an adsorption time (min). Co and C are initial and measured concentration, respectively.

Breakthrough capacity (q_b) was calculated using a breakthrough time (t_b) and the breakthrough time is measured when Kr/Xe is detected at first time in GC.

3. Adsorption Test

3.1 Adsorption Conditions

The commercial adsorbents, activated carbon and zeolite, and a novel adsorbent, polymer-based porous carbon, were selected as adsorbents.

In addition, Kr/Xe adsorption was tested with controlling parameters such as Kr/Xe concentration and adsorption temperatures. From the results, it was found that the adsorption capacities were highly influenced by the Kr/Xe concentration and adsorption temperature. The both breakthrough and equilibrium capacity increase as increasing Kr/Xe concentrations (Fig. 1 and 2), however, they decrease with increasing adsorption temperature.

Dynamic results were obtained through composition of gas using GC, and the adsorption performances such as working capacities, equilibrium capacity, and kinetic. Furthermore, the effects of adsorption condition on the adsorption capacities were investigated. These parameter effects will be further applied in optimizing adsorption process.

3.2 Adsorption performances

On the other hand, the feed gas in Pyroprocessing contains hydrogen or oxygen for reduction and oxidation, respectively, and therefore Kr/Xe adsorption test was carried out in 3.2% H_2-balanced with Ar or 21% O_2-balanced with Ar. It was confirmed that similar results were obtained in the presence of hydrogen or oxygen which indicate a feasible adsorption process in Pyroprocessing (Fig. 2).

4. Conclusion

Adsorption process was designed for Kr/Xe capture using packed bed column with adsorbents. Kr/Xe adsorption performance was investigated by varying adsorbents and process parameters. It was found that Kr/Xe adsorption was highly influenced by Kr/Xe concentration and adsorption temperature.

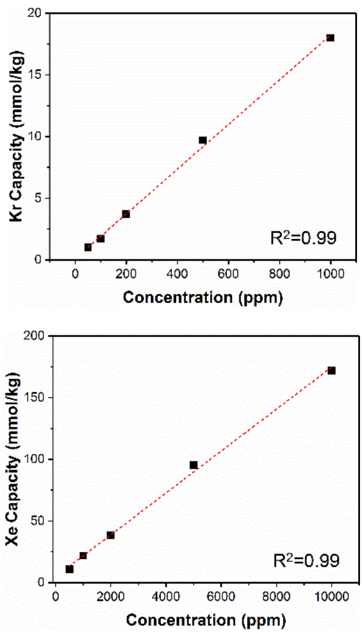

Fig. 1. Adsorption capacities at various Kr and Xe concentrations.

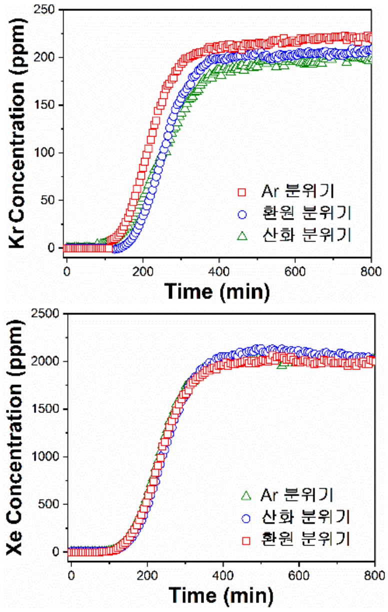

Fig. 2. Breakthrough curves measured at argon, reducing, and oxidizing condition.

REFERENCES

[1] L. Deliere, B. Coasne, S. Topin, C. Greau, C. Moulin, D. Farrusseng, “Beakthrough in Xenon capture and purification using adsorbent-supported silver nanoparticles”, Chemistry: A European Journal, 2, 9660–9666 (2016).

[2] M. Greenhalgh, T. G. Garn, J. D. Law, “Development of a hydrogen mordenite sorbent for the capture of krypton from used nuclear fuel reprocessing off-gas streams”, Journal of Nuclear Science and Technology, 51, 476481 (2014).