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Separation of Radioactive Xenon and Krypton from Off-Gas Streams Using PVDF-derived Porous Carbon

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1. Introduction

Used nuclear fuel (UNF) is a valuable and recyclable resource. However, the processing of UNF generates a stream of gaseous radioactive byproducts, often called off-gases, which include the noble gases xenon (Xe) and krypton (Kr) [1]. Effectively separating these radioactive gases from the off-gas stream is critical for ensuring both public safety and environmental protection. While various gas separation technologies exist, the adsorption method using porous carbon offers significant advantages in terms of cost and efficiency [2]. This research explores the separation of Xe and Kr using a porous carbon material synthesized from polyvinylidene fluoride (PVDF).

2. Methods and Results

2.1 Preparation of porous carbon adsorbent

Porous carbon adsorbents were prepared through the thermal treatment of polyvinylidene fluoride (PVDF) powder. The PVDF precursor was placed in an alumina crucible inside a tube furnace, and a continuous flow of high-purity argon gas was maintained throughout the process to prevent oxidation. The samples were heated at a rate of 10 °C/min from room temperature to a final carbonization temperature, which was then held for 2 hours. To investigate the effect of heat treatment temperature on the adsorbent's properties, six different samples were synthesized at temperatures ranging from 500 °C to 750 °C, with intervals of 50 °C. Each resulting sample was designated as CPPX, where CPP stands for Carbon from Pyrolyzed PVDF, and X represents the heat treatment temperature. After the thermal treatment was complete, the prepared CPPX samples were allowed to cool down to room temperature before being collected for further analysis.

2.2 Xe and Kr adsorption experiment

The adsorption experiments for Xe and Kr were conducted using a 3Flex (Micromeritics). Prior to each measurement, approximately 0.2 mg of the porous carbon adsorbent was degassed at a temperature of 350 °C for 24 hours under vacuum to remove any pre-adsorbed impurities. Static volumetric adsorption isotherms were measured for both Xe and Kr at three different temperatures: 10 °C, 25 °C, and 40 °C. The adsorption capacity was determined by measuring the amount of gas adsorbed by the sample at various pressures. All experiments were performed to a maximum pressure of 1 bar. The obtained adsorption data was used to evaluate the adsorbent's performance under different temperature conditions.

2.3 Adsorption performance of adsorbent

Table 1 shows the adsorption capacities of all six samples (CPP500 to CPP750) for both Xe and Kr at three different temperatures (10 °C, 25 °C, and 40 °C). Evaluating adsorption performance at such low pressures is crucial for developing adsorbents capable of capturing even trace amounts of radioactive gases, which is essential for effective off-gas treatment. The results indicate that the adsorption capacity for both gases increases as the carbonization temperature rises. The CPP750 sample, which was heat-treated at the highest temperature of 750 °C, consistently showed the highest adsorption performance, demonstrating its superior porous structure for gas capture. The adsorption capacity for both Xe and Kr decreases as the temperature increases. This is a typical characteristic of physical adsorption, as higher temperatures provide more kinetic energy to the gas molecules, weakening the physical bonds between the adsorbent surface and the gas molecules, thus hindering their adsorption.

Table 1: Adsorption Capacities of Porous Carbon Samples for Xe and Kr at 0.001 bar

Sample	Adsorption capacity at 10^{-3} bar (mol/kg)					
	Xe			Kr		
	10°C	25°C	40°C	10°C	25°C	40°C
CPP500	0.05	0.02	0.01	0.001	-	-
CPP550	0.08	0.05	0.01	0.003	0.001	-
CPP600	0.11	0.07	0.04	0.004	0.002	0.001
CPP650	0.13	0.08	0.05	0.005	0.003	0.001
CPP700	0.13	0.08	0.05	0.005	0.003	0.001
CPP750	0.16	0.11	0.07	0.006	0.004	0.002

Figure 1 specifically presents the adsorption isotherms of the best-performing sample, CPP750, for Xe and Kr at 10 °C, 25 °C, and 40 °C. The curves clearly show that the Xe adsorption capacity is substantially greater than the Kr capacity, reinforcing the potential of this adsorbent for selective Xe/Kr separation.

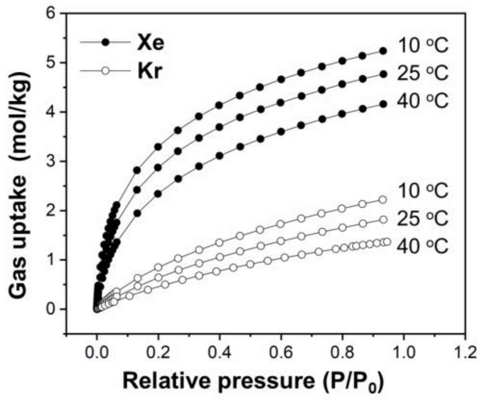

Fig. 1. Adsorption Isotherms of the CPP750 Sample for Xe and Kr at 10, 25, and 40 °C

As shown in Table 1 and Figure 1, the adsorption performance for Xe is significantly higher than that for Kr across all temperatures and samples. This remarkable difference in adsorption capacity is attributed to the larger polarizability of the Xe atom compared to the Kr atom. The stronger van der Waals forces between the porous carbon surface and the Xe atoms result in much higher adsorption capacities. This high selectivity confirms that the synthesized porous carbon is a suitable material for separating Xe from Kr.

3. Conclusions

In this study, we successfully prepared a series of porous carbon adsorbents (CPPX) by the thermal treatment of polyvinylidene fluoride (PVDF) for the selective separation of radioactive xenon (Xe) and krypton (Kr). The results confirmed that the carbonization temperature plays a crucial role in determining the material's adsorption performance. The CPP750 sample, heat-treated at the highest temperature, exhibited the most superior adsorption capacity for both Xe and Kr.

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