OPTIMIZING THE ENVIRONMENTAL FACTORS FOR THE AFFINITY OF 222Rn WITH CARBON FIBERS

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Prejem rokopisa – received: 2021-04-21; sprejem za objavo – accepted for publication: 2021-05-04

Radon (222Rn) is a member of the decay chain of 238U. It has been well documented that 222Rn is known to present a risk of lung cancer once inhaled. For practical implementation, this study makes use of activated carbon fibers (ACFs) to uptake 222Rn through environmental variations in the temperature, dosage of the adsorbent and circulate conditions. Here, three ACF samples were prepared via different carbonization and activation temperatures from 850 °C to 900 °C. The observed adsorption characteristics show that low temperature and high surface contact of an adsorbent with rich micropores allow the maximum adsorption of 222Rn. No such correlation was observed in the case of increasing the circulate conditions. However, there was a direct influence on the maximum adsorption capacity once the circulate conditions decreased. In summary, these findings expand the understanding of the adsorption, dependency on the factors and a proper carbonaceous-material design for reducing 222Rn.

Keywords: radon, carbon fibers, temperature, surface contact, pressure, flow rate

1 INTRODUCTION

Radon (222Rn) is a noble radioactive gas that emanates from the decay of 238U with a half-life of less than 4 days. The infiltration of 222Rn with its high mobility is considered to be the most significant cause of indoor radon exposure.1 Epidemiological studies have shown that residential radon exposure may cause lung cancer even at low radon levels.2–5 In this sense, an efficient uptake of 222Rn becomes quintessential. The use of activated carbons was first introduced in 1906 by Rutherford and flourished for decades in response to the above need.6 Activated carbon is widely applied in both gas and liquid-phase adsorbents due to its relatively low cost, high porosity and electric independence. However, adsorbate 222Rn molecules first need to pass through macro- and mesopore regions before entering micropore regions because of their ladder-like structure. Due to the natural promiscuity of activated carbon, activated carbon fibers (ACFs) can be an excellent alternative adsorbent since microporous characteristics allow 222Rn to bypass macro- and mesopore regions and directly reach micropore regions.7–10 Using ACFs can also be ideal for minimizing the waste disposal during the material synthesis by using the pitch from the oil-industrial sector as the precursor. Inspired by the above encouraging characteristics, we synthesized pitch-based ACFs as an adsorbent for the 222Rn adsorption. Here, surface characteristics of samples, including temperatures and circulate conditions, were determined as experimental variations. While previous studies merely observed the affinity of 222Rn with carbonaceous materials via surface areas, our work further explores the trinity of environmental factors for a better understanding.11 These findings may advance the fundamental knowledge of the 222Rn uptake using carbonaceous materials for an efficient design of adsorbents.

2 EXPERIMENTAL PART

Pyrolysis fuel oil was kindly supplied by Hyundai Oilbank and used as the precursor. The synthesis of ACFs was prepared using pyrolysis fuel oil. First, the temperature was elevated from room temperature to 300 °C under N2 atmosphere (2 L/min) for 1 h to remove
all the unnecessary impurities. Afterward, the material was spun in a centrifugal spinneret at a temperature of 245 °C. The resulting carbon fibers were then stabilized in air at 260 °C for another 5 h. For the carbonization and activation, carbon fibers were heated under N₂ (4 L/min) and H₂O atmosphere (0.2 mL/min) for 1 h, respectively, in a tubular furnace. Herein, temperature ranges from (850, 875 and 900) °C were designated for carbonization and activation to observe the correlation between the specific surface of samples and the adsorption capacity. We further denoted the samples by their carbonization and activation temperatures: ACF850, ACF875 and ACF900 for convenience.

The ²²²Rn measurement was conducted using passivated implanted planar silicon sensors adapted to RAD-7 (Durridge) at room temperature for 48 h. As illustrated in Figure 1, a parent nuclide (²²⁶Ra) was introduced to lure ²²²Rn inside the chamber. Each sample was heated at 90 °C for 3 h before the ²²²Rn measurement and the inner-atmosphere was circulated by designated conditions (a pressure of -0.02 – 0.02 MPa and a flow rate of 1–5 L/min). Physical characteristics of the samples were determined using ASAP 2460 (Micromeritics). The specific surface area (S_BET) was calculated with the Brunauer-Emmett-Teller (BET) method. The micropore area (S_M) and the external area (S_E) were evaluated with the Barrett-Joyner-Halenda (BJH) method using a N₂ adsorption isotherm. A CHN/S analyzer was employed to determine the contents of carbon, hydrogen and nitrogen using 2400 I (Perkin Elmer). Here, the oxygen content was calculated on the basis of the difference between the carbon, hydrogen and nitrogen contents. Scanning-electron-microscope images were observed under an accelerating voltage of 20 kV using S-3000H (Hitachi).

3 RESULTS AND DISCUSSION

Tables 1 and 2 list the properties of the samples. As the temperature increased from 850 °C to 875 °C, there was a corresponding increase in both S_BET (21.96 %) and S_M (36.28 %). The dependency of S_BET and S_M on the temperature was reduced from 900 °C onwards. The maximum values for S_BET and S_M were 1653 m²·g⁻¹ and 1447 m²·g⁻¹ at 875 °C, respectively. In all the cases, carbonaceous samples in the fiber form revealed a decent amount of micropores. It is also worth pointing out that the decrease in the C- and H-contents after the activation could be attributed to the dehydrogenation of linear structures to form an aromatic carbon network (Figure 2a). O-contents, in other words, surface functional groups (viz., carboxylic, lactonic and phenolic) were in the order of ACF875 > ACF900 > ACF850. This may account for the high textual properties of ACF875, which allow further integration of O-contents before and/or even after the fabrication once the surface is exposed to air (Figure 2b). Irrespective of whether the N- or S-contents vary insignificantly, the aforementioned data set consistently suggests that ACF875 with the highest physicochemical properties may reveal the highest ²²²Rn adsorption capacity among the samples.

The adsorption capacity of ²²²Rn under various conditions of temperature and sample dosage was investigated each time under a pressure of -0.02 MPa and flow rate of 1 L/min, respectively (Figure 3). Given that a simulta-
neous removal has practical applicability, up to 48 h was suggested as the maximum time frame. Concomitantly, a natural decay plot is also included in dark blue. The variation in the temperature suggests that ACF850 and ACF900 showed a much faster 222Rn adsorption within 0–36 h, as exemplified by robust surface characteristics of meso- and macropores. After 36 h, while again no significant adsorption capacity was identified in ACF850 and ACF900, plots of ACF875 showed a drastic shift due to a continuous accumulation of 222Rn onto micropores. Based on the physicochemical properties, we assumed that ACF875 would have the most 222Rn adsorption capacity.

Calculating the values from Figure 3c, the final adsorption capacity was in the order of ACF875 (135.2 Bq/m³) > ACF900 (141.1 Bq/m³) > ACF850 (148.1 Bq/m³). This order can be found in relevant adsorption studies using porous media. The temperature was in an inversely proportional correlation with the adsorption capacity in all the cases except for ACF875. This is in accordance with the previous studies, suggesting that the 222Rn adsorption coefficient at zero temperature shows an exponential increase. This observation is also consistent with the variation in the sample dosage,
showing that ACF875 had a slow start but ended up with the highest adsorption capacity of 222Rn. According to the data acquired from Figure 3e, the final adsorption capacity was in the order of ACF875 (100.01 Bq/m³) > ACF900 (104.41 Bq/m³) > ACF850 (108.72 Bq/m³). The data from these experiments also allows us to compare the environments affecting the effective 222Rn adsorption.

Though the surface geology of ACFs first seemed to imbibe 222Rn via micropores, different proportions of micropores of each sample resulted in an unpredictable adsorption capacity within a long-term scenario. It should also be noted that the sample dosage showed a greater influence than the temperature in all the cases (Figure 3f). Given that the variation in the sample dosage may be insufficient for the 222Rn adsorption capacity over 50 %, a chemical modification of the surface or the use of other material-based adsorbents may further increase the numerical values of adsorption.

According to the relevant literature, circulate pressure is in a linear correlation with the adsorption capacity. In the case of 222Rn, a decrease in the circulate pressure may enhance the overall flux as shown by early experiments. A comparison of the 222Rn adsorption capacity with 20-g samples at a flow rate of 1 L/min over 48 h is summarized in Figure 4a. The adsorbed values of 222Rn at a low circulate pressure appear to be high because of the enhanced flux. The observed value of ACF875 (114.63 Bq/m³) at –0.02 MPa is comparable to that of ACF900 (119.95 Bq/m³), but the values in the cases of ACF850 (131.56 Bq/m³) and natural decay (160.34 Bq/m³) are higher. Likewise, the 222Rn adsorption is higher at a low flow rate (–0.02 MPa) compared to the values of –0.01–0.02 MPa (Figure 4b). This is mainly due to the competition between the surrounding radioactive species and the limited surface pores. From the above results, it can be concluded that all the prepared samples show increased values of 222Rn adsorption capacity at low temperatures, high contact surfaces, low pressures and high flow rates.

4 CONCLUSION

It is clear from the results of our experiments that a combination of factors including the temperature, contact surface and circulate conditions can effectively enhance the uptake of 222Rn. Comparable though less pronounced measures are also available for enhancing the adsorption capacity of 222Rn such as various chemical modifications of the surface of an adsorbent. In summary, with the above calculations and observations, we de facto identified the adsorption mechanisms of 222Rn and may continue our research in terms of an efficient adsorbent design.

Acknowledgements

This work was supported by the Korean Energy Technology Evaluation and Planning (Project No. 20171520000300).

Declaration of interest

The authors declare that there is no conflict of interest.

5 REFERENCES


