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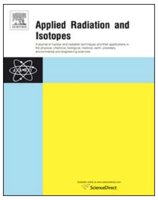

The temperature dependence of adsorption coefficients of ^{222}Rn on activated charcoal: an experimental study

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ABSTRACT

The radon adsorption coefficient of activated charcoal (K) has exponential relationship with temperature theoretically, but few experiment results of K at temperature below 0 ℃ were given. In this study, K were measured using a flow-through system with activated charcoal in cylindrical adsorption beds at temperature adjusted from room temperature to −48 °C using liquid nitrogen. Results are consistent with theory and show that the adsorption coefficient at −48 °C is nearly 25 times higher than that at 23 ℃.

1. Introduction

Activated charcoal has numerous applications in removing pollutants from air or water streams due to its porosity. In uranium mills and thorium processing facilities, activated charcoal is used as decay beds and removal system (Fusamura et al., 1963; Ackley, 1975). Activated charcoal canister is also widely used in radon concentration field survey due to its small, inexpensive and power supply independence (Bernard, 1983; George, 1984; Prichard and Marien, 1985). Recently, activated charcoal is used for radon removal in low background laboratory, such as Borexino (Italy)(Pocar, 2003), SNO+(Canada)(Golightly, 2008) and super-Kamiokande (Japan) (Fukuda et al., 2003).

Radon adsorption coefficient, which is defined as the ratio of radon activity on charcoal to radon concentration in air at equilibrium, is commonly used to describe the adsorption ability of activated charcoal. Radon adsorption coefficient not only depends on the properties of the activated charcoal (porosity, grain size), but also influenced by environmental factors such as air pressure, humidity and temperature (Ronca-Battista and Gray, 1988; Gaul and Underhill, 2005).

The temperature has great influence on the radon adsorption coefficient. When the temperature goes down, the adsorption coefficient goes up quickly. The theoretical relationship between radon adsorption coefficient and temperature can be expressed as follow (Shefsky et al., 1993):

$$K = K_0 e^{\frac{Q}{RT}} \tag{1}$$

Where, K_0 is of the nature of frequency factor (g/L), Q is the adsorption heat (J/mol), R is the molar gas constant 8.314 J/mol/K (1.987 cal/mol/K).

However, few measurement results of radon adsorption coefficient under different temperature were given before, especially at temperature below zero. Gübeli (Gübeli and Störi, 1954), Luetzelschwab (Luetzelschwab et al., 1994) and Samman (Samman et al., 2002) measured the radon adsorption coefficient at different temperature above 0 °C. Zikovsky (Zikovsky, 2001) and Golightly (Golightly, 2008) performed a few measurements at temperature below 0 ℃, but the experimental results were quite limited.

To study the temperature dependence of radon adsorption coefficients on activated charcoal, a series of experiments were carried out. The temperature of activated charcoal was adjusted from room temperature to −48 °C using liquid nitrogen. The radon adsorption coefficients at different temperature were measured using a flow-through system with activated charcoal in cylindrical adsorption beds.

2. Materials and methods

Due to its high porosity, the coconut shell activated charcoal KC-6[1] with mesh No. 10–20 was selected in this study. The radon adsorption coefficient of this activated charcoal is 10.4 g/L at 12 °C (Yunxiang, 2016). The activated carbon was dried at 120 °C for more than 48 h, and cooled to room temperature in a desiccant jar for at least 24 h. Then it was filled in polyethylene columns and used as an adsorption bed.

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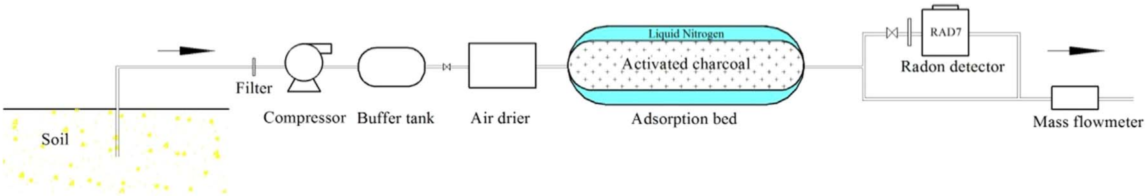

Fig. 1. The flow-through system for radon adsorption coefficient measurement.

Table 1
Experiment measurement of adsorption coefficient at different temperatures.

Temperature ℃	Temperature K	Mass of charcoal g	Flow rate L/min	Breakthrough time min	Adsorption coefficient L/g
23	296.15	181.0	8.3	150	6.9 ± 1.7
12	285.15				10.4 ± 1.6[*]
−2	271.15	90.4	18.0	95	18.9 ± 3.4
−18	255.15	14.6	28.0	27	51.8 ± 9.8
−33	240.15	15.0	32.5	47	102 ± 12
−44	229.15	5.1	46.0	17	153 ± 25
−48	225.15	4.2	45.0	16	171 ± 33

[*] Result at 12 °C is quoted from the former work (Yunxiang, 2016).

To measure radon adsorption coefficient of the activated charcoal at different temperature, a flow-through system with activated charcoal in cylindrical adsorption beds was set up, as it's shown in Fig. 1.

Radon gas from soil was drawn out by a compressor DA5002,[2] the follow rate could be controlled and the output pressure was nearly 1 atm. Then the gas went through an air drier DE0005[2] with dew point −40 °C, and entered the adsorption bed. The adsorption bed was surrounded by a layer of liquid nitrogen, where the temperature of charcoal was adjusted through liquid nitrogen. A temperature sensor L99-TWS-2[3] was inserted into the adsorption bed to monitor the temperature. The radon concentration in output airstream was measured by RAD7 radon monitor (Durridge, The USA) with a cycle of 2 min. The follow rate through the adsorption bed was measured by a mass follow-meter (TSI4046, TSI, The USA).

After continuously drawn into the activated charcoal column, Radon gas will come out in the downstream after a certain time delay. This delay time is referred as the breakthrough time (τ). According to the HETS model (Littlewood, 1970), the adsorption coefficient (K) can be calculated by measuring τ (min), air flow rate ϕ(L/min) and mass of activated charcoal m (g).

$$K = \frac{\tau \Phi}{m} \tag{2}$$

Actually, the measured radon concentration has a stretched S-shape curve, and the breakthrough time τ can be defined as the time when 50% saturation radon concentration is reached. This method is easy to operate when there is stable radon source, and very practicable in situation of radon removal in indoor environment (Karunakara et al., 2015; Xie et al., 2011). We used soil gas as a steady radon source with average radon concentration of 3600 $\mathrm{Bq\,m^{-3}}$. A probe was driven into the soil to a depth of more than 1 m, just outside the laboratory building, and a compressor was connected to the probe to draw the soil gas.

Considering the boiling point of radon is −61.7 °C Prichard (1987), adsorption coefficient measurements were performed at six different temperatures from room temperature down to −48 °C. According to theory evaluation, the adsorption coefficient would be much higher at low temperature. So we adjusted the mass of charcoal and flow rate to limit the breakthrough time.

3. Results and discussion

Six individual measurements were conducted for the adsorption coefficient at temperature from 23 °C to −48 °C. The experiment parameters and results are shown in Table 1.

It is significant from our results that the adsorption coefficient increases rapidly when the temperature goes down, and the adsorption coefficient value at −48 °C is nearly 25 times higher than the value at 23 °C.

Put the measured radon adsorption coefficients as well as former reported results into one picture, the results are shown in Fig. 2.

It can easily be seen that the radon adsorption coefficient decreases quickly with the temperature going up. Compared with the results of former researches, our result is consistent with them at temperature above 0 °C, while at temperature below 0 °C, the difference exists probably due to the different kinds of activated charcoal.

Fitting was made based on Eq. (1). The value of K_0 is 0.0032 ± 0.0029 L/g and Q/R is 2461 ± 203 K. As R is 8.314 J/mol/K, the adsorption heat Q of activated charcoal KC-6 can be estimated to be 20.5 ± 1.7 kJ/mol. This value is comparable to 18.8 kJ/mol, which Zikovsky estimated for activated charcoal from Fisher (5-685-A, 6–14 mesh) (Zikovsky, 2001), and lower than 30 kJ/mol derived by Golightly for charcoal from Norit (12 × 40 mesh) (Golightly, 2008).

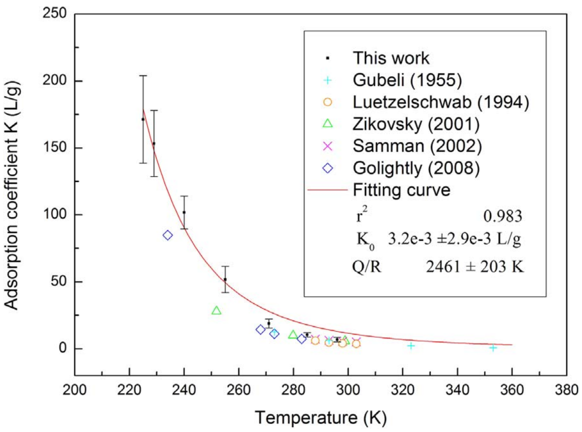

Fig. 2. Temperature dependence of adsorption coefficient of this and former studies.

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[2] Soret Gas Equipment (Beijing) Co., Ltd, Beijing, China.

[3] Hangzhou Luge Science & Technology Co., Ltd, Hangzhou, China.

4. Conclusion

For better understanding the temperature dependence of radon adsorption coefficient on activated charcoal, a series of experiments were performed at the temperature range from 23 °C to −48 °C.

Results show that adsorption coefficient increases exponentially when temperature decreases, which is consistent with theory. The adsorption coefficient of activated charcoal KC-6 at −48 °C, 171 L/g, is nearly 25 times higher than the value at 23 °C. At temperature above 0 °C, our results consist with former researches, while at subzero range the difference appears. The heat of adsorption of KC-6 is estimated to be 20.5 ± 1.7 kJ/mol, and it is within the range of former researches' results.

When used for radon removal, activated charcoal at low temperature will remarkably increases the efficiency and reduces the volume of activated charcoal needed, which is quite important for some special environment, such as low background laboratory.

Besides, when temperature is lower than the boiling point of radon −61.7 °C, radon is condensed in the adsorption bed, which means the radon adsorption coefficient is infinite, which is quite interesting.

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