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Measuring and understanding radon adsorption in microporous materials

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On behalf of SuperNEMO collaboration

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Abstract. The background from the radon decay chain is the strongest constraint for many experiments working at low energy and very low counting rate. A facility for studying the optimum radon capture by very selective porous materials was developed at CPPM in the context of the SuperNEMO project. In collaboration with Institut Jean Lamour, studies were carried out for better understanding radon adsorption in carbon adsorbents.

INTRODUCTION

Radon is one of the most problematic radioactive gases for low energy and low counting experiments. It is a noble gas which belongs to the U and Th chains. Radon has typically high mobility, allowing easy escape from bulk materials and diffusion into the active parts of detectors. Later on, through successive decays, it can induce an important background in low energy physics experiments. For example, in SuperNEMO double beta decay project [1], the concentration of radon in the detector gas has to be below 150 $\mu\text{Bq}/\text{m}^3$. To reach such quite a low concentration, purifying the gas detector is required.

The only practical possibility to capture radon from a carrier gas is based on physisorption, caused by van der Waals forces at the surface of materials. However, the adsorption phenomenon corresponds to a dynamic equilibrium, as radon atoms continuously adsorb onto, and desorb from, the material's surface. The capture of radon is therefore not everlasting as its evolution from the adsorbent is only slowed down, however with a significantly decreased equilibrium partial pressure. The elapsed time between introduction and detection of radon at both ends of an adsorbent column is called "retention time". If the retention time is much larger than radon lifetime, the radon capture can be considered as complete. Temperature, pressure, adsorbent's surface area and pore-size distribution, as well as competitive adsorption of radon and carrier gas, are important parameters to be considered for the optimization of radon capture on any given material.

Activated charcoal is a well-known adsorbent, widely used for the capture of various gases [2] [3]. Activated charcoal is a highly disordered solid, presenting a quite high internal surface area and a rather broad pore-size distribution, ranging from micropores (< 2 nm) to mesopores (2 – 50 nm). In contrast, several synthetic microporous carbons materials such as carbon molecular sieves (CMS), having a much narrower pore-size distribution and hence a much higher expected selectivity with respect to radon atoms, were developed for chromatography and atmospheric chemistry applications. CMS might therefore be more suitable than activated charcoal for adsorbing radon.

In the context of the SuperNEMO collaboration, a test bench was developed at CPPM to study radon adsorption onto various microporous materials (activated charcoals, carbon molecular sieves, Metal Organic Frameworks, ...), presenting a great ability to capture radon from different carrier gases in different experimental conditions.

EXPERIMENTAL SETUP

The experimental setup is shown in Figure 1. Low amounts of radon were introduced in the carrier gas (nitrogen) by using as radon source a metal plate coated with a thin radium layer, maintained at a fixed temperature. The gas was then introduced in a buffer tank in which radon concentration, temperature, pressure and relative humidity were measured. Thereafter the carrier gas, containing a well-defined amount of radon, was introduced in the column trap located into a freezer. The saturation of the trap was measured with a commercial RAD7 radon detector, calibrated for a continuous nitrogen flow. All measurements were carried out with a flow of 10 L/h, a pressure of 1 bar and a mean radon activity of $880 \pm 23 \text{ Bq/m}^3$. The activity of the saturated column was measured by gamma spectrometry with a Ge detector using the main gamma lines of ^{214}Pb and ^{214}Bi [4].

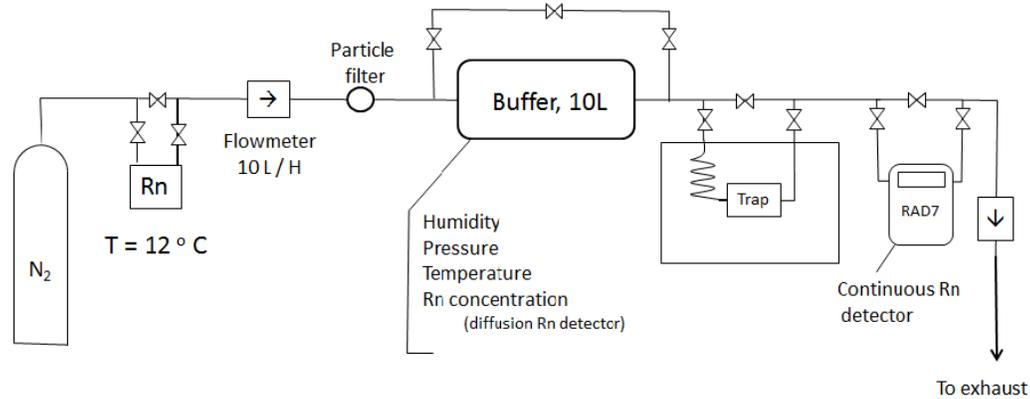


FIGURE 1. Experimental setup.

The equilibrium was assumed to be reached between radon flowing in the carrier gas and radon adsorbed in the trap when the concentration at the outlet of the trap was constant. In such conditions, the ratio of the number of radon atoms trapped to that remaining in the gas phase, both proportional to their respective activity, led to the equilibrium constant K :

$$K = \frac{\{A\}}{\{C\}} [m^3/kg]$$

where $\{A\}$ is the radon activity per unit mass (Bq/m^3) in the trap measured by the Ge detector, and $\{C\}$ is the radon activity in the gas (Bq/m^3) measured by the diffusion radon detector.

ADSORBENT MATERIALS TESTED

Only adsorbents having high surface areas and relatively low specific radioactivities were considered in the present work. Nevertheless, in order to better understand the radon adsorption phenomenon, studying standard zeolites and other materials also presenting a great potential for radon capture is foreseen in the near future, even if they present a too high intrinsic radioactivity for the SuperNEMO experiment.

The materials selected for this preliminary study were, see Table I:

- classical activated charcoals (K48 special¹, K48¹, NuclearCarb², Envirocarb², G2X4³), characterized by high surface areas and broad pore-size distributions;

¹ Classical activated carbon from Sillicarbon

² Classical activated carbon from Chemviron Carbon

³ Classical activated carbon from Japan EnviroChemicals

- carbon molecular sieves (Carbosieve SIII⁴, Carboxen 1000⁴, and 569⁴), characterized by well-defined, narrower, pore sizes;
- synthetic carbon (Carboact⁵), widely used in low background experiments.

TABLE I. Radon adsorption K-factor parameter for several adsorbents investigated at 20°C, 0°C, -30°C, and -50°C.

Sample	K factor at 20°C (m ³ /Kg)	K factor at 0°C (m ³ /Kg)	K factor at - 30°C (m ³ /Kg)	K factor at - 50°C (m ³ /Kg)
K48	13 ± 1.5	14 ± 1.5	77 ± 6.9	232 ± 20
K48S	11 ± 1.2	19 ± 2.0	69 ± 6.1	276 ± 23
G2X4	6.3 ± 0.7	24 ± 2.6	69 ± 6.1	253 ± 22
NUCLEARCARB 208C 5KI3	7 ± 0.8	15 ± 1.6	55 ± 4.9	190 ± 16
NUCLEARCARB 208C 5TEDA	3.9 ± 0.4	11 ± 1.2	40 ± 3.6	170 ± 14
ENVIRONCARB 207C	13 ± 1.5	21 ± 2.2	101 ± 9.0	270 ± 23
Carboact	17.1 ± 1.9	36 ± 3.9	182 ± 16.2	480 ± 41
carboxen 1000	11.4 ± 1.3	35 ± 3.7	148 ± 13.2	484 ± 41
carboxen 569	2.2 ± 0.2	7.8 ± 0.8	14 ± 1.2	100 ± 9
carbosieve SIII	16.5 ± 1.9	68 ± 7.3	160 ± 14.2	518 ± 44

Table 1 shows the K-factor for ten samples measured at 20°C, 0°C, -30°C and -50°C. As it can be seen, carbon molecular sieves (especially Carbosieve III and Carboxen 1000) presented a high affinity for radon at low temperatures (-50°C), much higher than that of standard charcoals. This advantage, however, decreased when the temperature increased, so that the adsorption capacity of carbon molecular sieves at room temperature was similar to that of the best activated charcoals.

ADSORPTION ANALYSIS

A series of charcoals and carbon molecular sieves were investigated at Institut Jean Lamour. N₂ and CO₂ adsorption-desorption isotherms were measured for each material. From these data, the corresponding pore-size distributions (PSDs) were obtained. As an example, Figure 2 shows the PSDs of K48 charcoal and Carbosieve SIII samples. As expected, CMSs have narrower PSDs than regular charcoals.

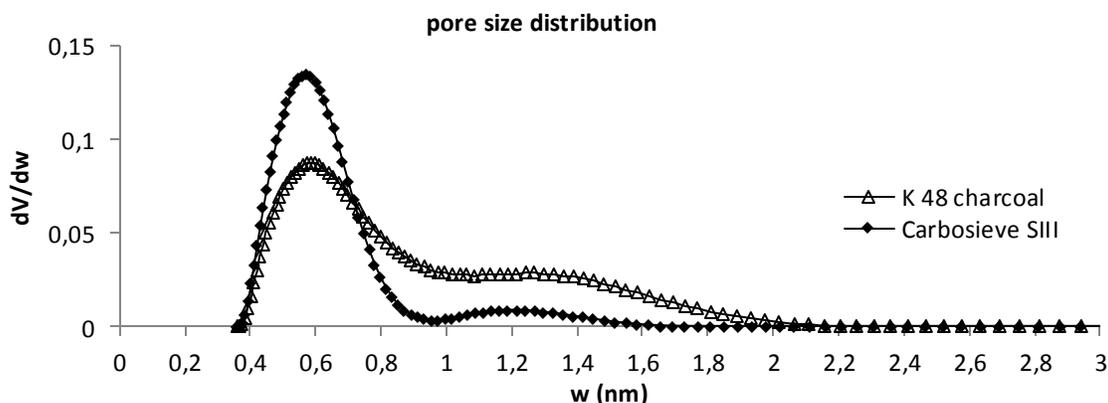


FIGURE 2. Pore size distributions of a standard charcoal and a carbon molecular sieve.

⁴ Carbon Molecular Sieve from Supelco, Inc.

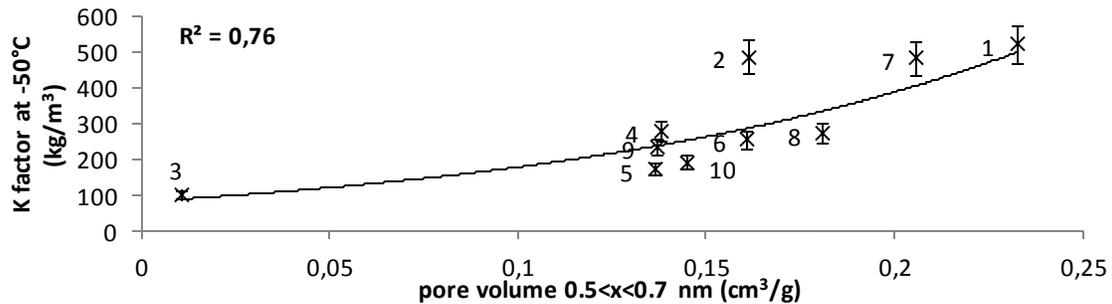
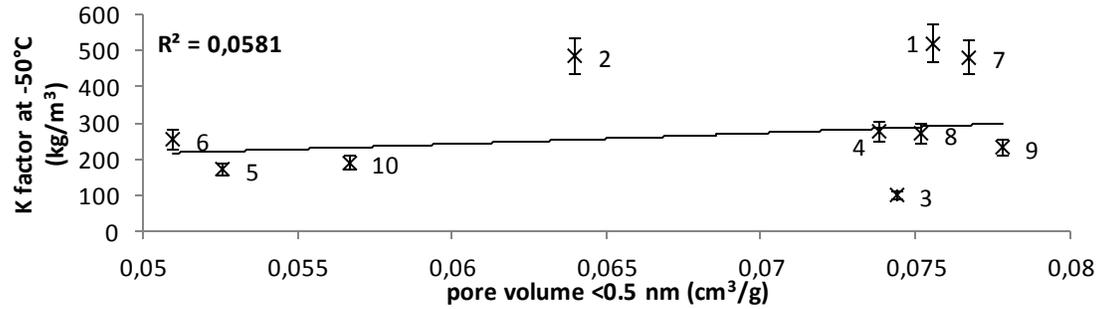
⁵ Synthetic activated charcoal from Carboact

The PSDs were used to calculate the cumulated pore volume in several pore size intervals: < 0.5 nm, between 0.5 and 0.7 nm, < 0.7 nm (ultramicropores), between 0.7 and 2 nm (supermicropores), < 2 nm (micropores) and above 2 nm (mesopores).see table II

TABLE II. Textural analysis of material used

Sample	Surface area BET (m ² /g)	Pore volume <0.5 nm (cm ³ /g)	pore volume <0.7 nm (cm ³ /g)	pore volume 0.5-0.7 nm (cm ³ /g)	pore volume 0.7-2 nm (cm ³ /g)	pore volume <2 nm (cm ³ /g)
K48	793	0.078	0.215	0.137	0.099	0.314
K48S	799	0.074	0.212	0.138	0.105	0.317
G2X4	1383	0.051	0.212	0.161	0.322	0.534
NUCLEARCARB 5KI3	1315	0.057	0.202	0.145	0.321	0.522
NUCLEARCARB 5TEDA	1297	0.053	0.189	0.137	0.321	0.510
ENVIRONCARB 207C	1073	0.075	0.256	0.181	0.165	0.421
Carboact	1096	0.077	0.283	0.206	0.156	0.439
carboxen 1000	812	0.064	0.225	0.161	0.045	0.270
carboxen 569	299	0.074	0.085	0.011	0.000	0.085
carbosieve SIII	1061	0.076	0.308	0.233	0.102	0.410

The objective was to find a relationship between pore diameter and radon adsorption. Thus, the K-factor was plotted as a function of pore volume for each range of considered pore diameters.



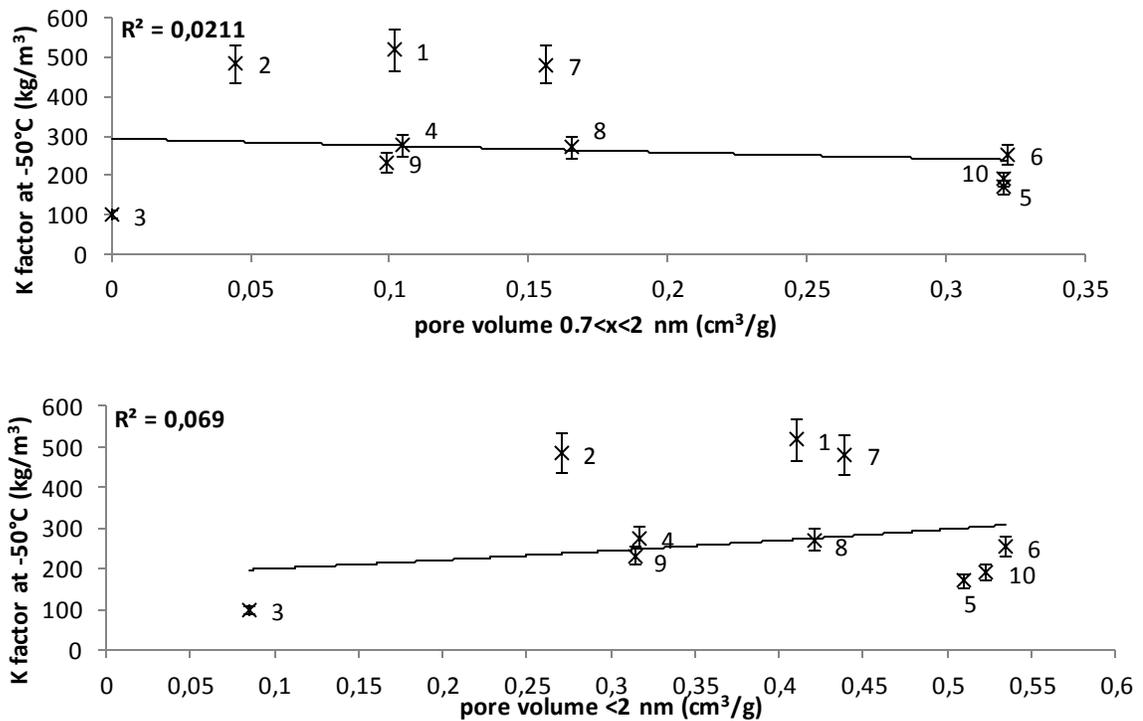


FIGURE 3. Radon adsorption K-factor measured at -50°C , as a function of cumulated pore volume (for pore diameters below 0.5 nm, ranging from 0.5 to 0.7 nm, ranging from 0.7 to 2 nm and below 2 nm), for several samples (1: Carbosieve III; 2: Carboxen 1000; 3: Carboxen 569; 4: K48S; 5: Nuclecarb 208C-5TEDA; 6: G2X4; 7: Carboact; 8: environcarb_207; 9: K48; 10: Nuclecarb 208C 5KI3). The lines are exponential fits to the experimental data.

The micropore volume (< 2 nm) was not found to be correlated with the K-factor. But a clear trend between K-factor and cumulated volume of pores with diameters ranging from 0.5 to 0.7 nm was observed, as shown in Figure 3. The PSD thus seems to be an important parameter for selecting materials having the highest radon adsorption capacity. Except for a few samples, the values of K-factor were indeed correctly fitted by an exponential law, R^2 being the correlation parameter. The same correlation was always seen, whatever the adsorption temperature. This finding suggests that radon is preferentially adsorbed in this range of pores, and that physisorption is the main contribution to radon adsorption. However, heteroatoms, ashes or surface functionalities might also affect radon-carbon interactions and explain deviations.

CONCLUSION

In the context of the SuperNEMO collaboration, a facility to study and optimize radon adsorption on porous materials was built. Several measurements based on commercial microporous carbon materials were carried out, from which very important information on the radon adsorption process was obtained. A better understanding of radon adsorption properties of carbon materials is now to be expected. Extending our work to different carrier gases is also planned in the near future.

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