PARAMETERS FOR CHARACTERIZATION OF RADON ADSORPTION AND DIFFUSION IN POROUS MEDIA

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ABSTRACT

Theoretical models and experimental methods are described for the determination of parameters used in the analysis of Radon adsorption and diffusion in porous media.

INTRODUCTION

Adsorption on activated charcoal is a phenomenon widely used for the measurement of Radon concentration in air.[1] Following exposure to an unknown atmosphere, the Radon content in a charcoal canister is derived from the γ-emission of its decay products, at radioactive equilibrium. Radon concentration in the air is then usually inferred by comparing the above mentioned γ-activity to the activity resulting from exposure to a known Radon concentration.

The Nuclear Instrumentation and Measurements Laboratory of DCMN is involved in the definition of the Italian standard for Radon measurement by means of activated charcoal. Among the goals of this effort is the identification of simple and accurate procedures for the determination of two fundamental parameters governing Radon permeation in porous media: adsorption and diffusion coefficients.

THEORETICAL MODELS AND EXPERIMENTAL MEASUREMENTS

Adsorption coefficient. This coefficient is found by injecting a gas pulse into a charcoal column open at both ends and analyzing the temporal evolution of the gas concentration at the outlet.[2,3]

A mathematical description of the pulse shape through the porous medium is given by the "theoretical plate model," borrowed from the analysis of plate distillation columns, which treats adsorption as a discrete process. The adsorbing column is regarded as a series of N chambers of height H where immediate equilibrium is reached between mobile and stationary phases. The pulse shape through the filter can be deduced, at any time t, from a mass balance, setting an initial condition of zero concentration in all the plates but the first one, where the input pulse is assumed as uniformly distributed. This yields:

$$C_N(t) = A \cdot \frac{N^N \cdot Q^{N-1} \cdot t^{N-1}}{(N-1)! \cdot (k \cdot m)^N} e^{-(N \cdot Q \cdot t / k \cdot m)}$$

The gas concentration at the outlet, calculated from this expression, is:

$$C_N(t) = \left(\frac{N \cdot Q \cdot t}{k \cdot m}\right)^{N-1} \cdot e^{-(N \cdot Q \cdot t / k \cdot m)}$$

The time t_{MAX}, at which the output concentration is maximum, and the time interval $\Delta t_{1/2}$, (FWMH), may be easily derived from experimental eluition curves (Figure 1). The number N of the theoretical plates may be then deduced from the relation between $\Delta t_{1/2}/t_{MAX}$ and N, reported in Figure 2. The relation between t_{MAX} and the average retention time, $\bar{t}$, is: $t_{MAX} = \bar{t} \cdot (N-1)/N$.

Finally, the adsorption coefficient K, relative to a column containing a charcoal mass m and crossed by an air flow Q, may be calculated from the expression: $K = (\bar{t} \cdot Q/m)$.

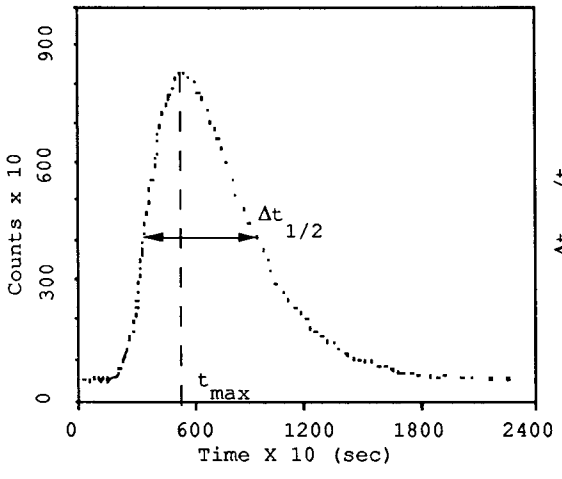

Figure 1. Eluition curve

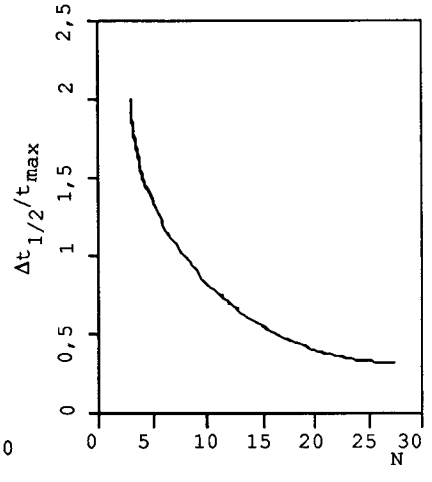

Figure 2. $\Delta t_{1/2}/t_{max}$ as a function of N

Effective diffusion coefficient. In order to measure the coefficient D_e, a charcoal column with one open end is exposed to a Radon atmosphere until a uniform concentration C_0 is reached throughout its height, a. A continuous flow of air then sets at zero the upper free surface concentration, while the γ-emission from Rn daughters is monitored with time. With these initial and boundary conditions, integration of the set of differential equations describing the process of combined eluition and decay

$$\frac{\partial C}{\partial x} = D_e \frac{\partial^2 C}{\partial t^2} - \lambda C$$

$$C(0,x) = 0$$

$$C(t,a) = 0$$

$$\left[\frac{\partial C}{\partial x}\right]_{t.x=0} = 0$$

yields a concentration profile along the column axis which approaches a single sinusoid when $t > \frac{1}{2\ D_e} (\frac{a}{\pi})^2$.

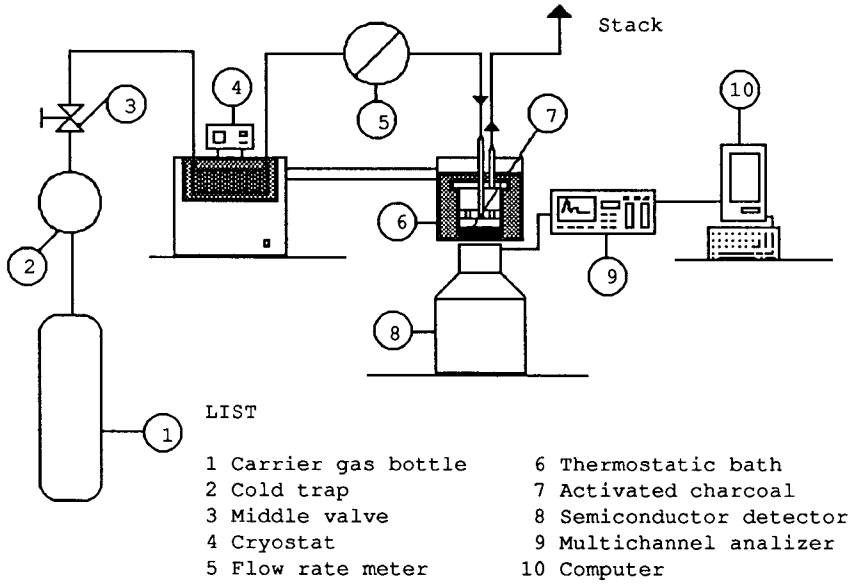

Figure 3. Test-rig

In this case, the distribution of radioactive isotopes inside the charcoal column does not vary with time, therefore, the geometric efficiency of a γ-detector placed near the column is constant. This allows for the measurement of the decrease of activity:[4]

$$A(t) = \frac{8\ C_0\ a}{\pi^2}\ e^{-\left[D_e \left(\frac{\pi}{2a}\right)^2 + \lambda\right] t} \qquad t > \frac{1}{2 \cdot D_e} \left(\frac{a}{\pi}\right)^2$$

The value of the constant $\left[D_e \left(\frac{\pi}{2a}\right)^2 + \lambda\right]$, where the diffusion coefficient D_e is the only unknown quantity, may be calculated through exponential interpolation of the experimental data, as shown in Figure 4.

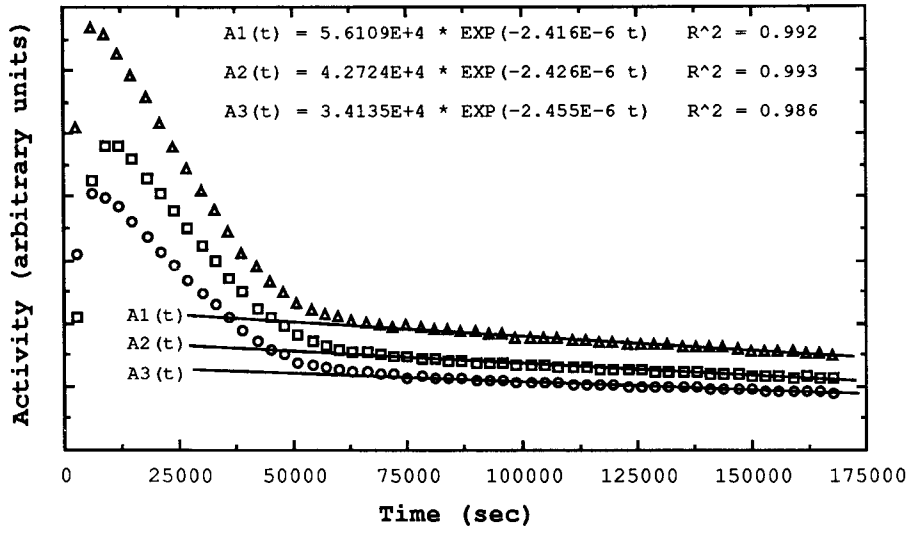

Figure 4. Temporal evolution of the γ-activity of the three most pronounced peaks of the Radon daughters spectrum.

CONCLUSIONS

At DCMN we can measure all the quantities that characterize the response of an activated charcoal canister exposed to constant conditions until equilibrium is reached with both environmental humidity and Radon. Based on these data, we are developing the analysis of the canister response when conditions of exposure undergo dynamic transients.

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