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Adsorption of Radon from a Humid Atmosphere on Activated Carbon

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Abstract

Temperature and relative humidity can influence the adsorption capacity of radon on activated carbon to a great extent, depending on the physical properties of the carbon. Experiments were carried out to measure the radon uptake by an activated carbon in the presence of water vapor in a specially designed adsorption apparatus. The radon concentrations in the gas and solid phases were measured simultaneously once the adsorption equilibrium and the radioactive equilibrium between the radon daughter products were reached. The experiments in the presence of water vapor were carried out using two approaches. In one case the activated carbon was preequilibrated with water vapor prior to exposing it to radon. In the other case the carbon was exposed to a mixture of water vapor and radon. The uptake capacity for radon decreased substantially when both components were introduced together compared to when carbon was preequilibrated with water.

INTRODUCTION

Radon is a radioactive gas produced by the decay of naturally occurring radium. It is present in soils, groundwater, and in various building materials such as brick and concrete. It enters a building from the soil beneath the structure through cracks and joints, such as in the structure foundation, through slabs and walls, floor drains, sumps, and through openings around utility lines. Radon generated within the earth's crust can dissolve in the groundwater and result in concentrations typically in the range of several thousand picocuries per liter. It can diffuse into the air from water and contribute to indoor radon concentrations (*6*). Similarly, building materials of natural origin, such as clay brick, marble, and sandstone, have a wide

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range of radium contents, and indoor radon levels can increase by a factor of 2 to 3 when air passes through these materials. The U.S. Environmental Protection Agency has estimated that radon concentrations in several million residences in the United States are significantly higher than the recommended level of 4 pCi/L and, as a major indoor air pollutant, they pose a serious health threat to occupants.

The most common and inexpensive method to measure radon concentrations indoors is by using carbon canisters. Rutherford (*15*) observed that activated carbon has the ability to adsorb radon from the gas phase and retain it in its pores. Activated carbon in canisters is now widely used and is recommended for indoor radon measurements by the EPA, as well as by numerous other public agencies and private companies.

The canister, which is impervious to radon diffusion, is exposed to indoor air which contains radon for a specified period of time. During this time period, radon is allowed to adsorb passively onto the surface of the charcoal. The canister is then sealed for future radon analysis. The gamma activities of radon daughter products, particularly ^{214}Pb and ^{214}Bi, are measured by a gamma ray spectrometer using an NaI(Tl) scintillation detector. The corresponding radon concentration in indoor air is then calculated from the activity, after correcting for the efficiency of the radon detector and the decay of radon with time. The effect of relative humidity on the adsorption capacity is taken into account by using a "calibration factor" that has a dimension of liters per minute. The error in radon measurements made by this method can be as high as 133%, with an average error of ±19% (*20*).

Although most researchers agree that the adsorption efficiency of the canisters decreases with an increase in temperature and humidity, disagreement exists over the extent to which these factors affect the measurements. Cohen (*2*) observed that the correction due to the change in relative humidity was not more than 6%. However, George (*5*), Ren and Lin (*13*), Scarpitta and Harley (*16*), and Ronca-Battista and Gray (*14*) found from their experiments that the calibration factor for relative humidity decreased by as much as 50% when humidities changed from 20 to 100%. Interestingly, Ren and Lin (*13*) and George (*5*) reported that the amount of radon adsorbed by canisters did not change significantly when temperature was changed from 17 to 27°C. On the other hand, Cohen (*2*) observed a change of 1.5% in adsorption capacity for each 1°F change in temperature.

Scarpitta and Harley (*16*) investigated the adsorptive and desorptive characteristics of canisters that contained a petroleum-based charcoal, Witco 6–12 mesh, under controlled conditions of temperature, relative

humidity, and radon concentration. Two sets of experiments were conducted. In the first, the charcoal was spread in a layer, which the authors termed a "monlayer." In the second, a packed bed of charcoal was used. The exposure time in both cases varied from 1 to 7 days. The results demonstrated that radon adsorption and desorption are dependent on the bed depth and on the amount of water adsorbed. The adsorption of radon decreased by one order of magnitude when water started condensing in the pores of the charcoal. Conventional charcoal canisters can become saturated in less than 4 days at 70% relative humidity if exposed in the fully opened configuration.

In order to improve the reliability of radon measurements by charcoal canisters and to extend the sampling period, a diffusion barrier is placed between the charcoal and the ambient air being sampled (*3*, *11*). The diffusion element can make the canister nearly independent of the properties of different charcoals and less sensitive to changes in radon concentrations and relative humidities, provided a longer integration time—up to 1 week—is used (*5*). Cohen and Nason (*3*) developed their own charcoal canister that employed a diffusion barrier. The canister dimensions were optimized by solving diffusion equations using a computer model. The humidity problem was avoided by placing a bag filled with a desiccant material inside the diffusion barrier. The authors indicated that this type of canister can be used for exposure times as long as 1 week without loss of significant accuracy in the measurement.

Pojer et al. (*10*) assessed the effects of temperature and humidity, both theoretically and experimentally, on the performance of a diffusion barrier charcoal canister. The adsorption capacity decreased by 30% when the temperature was increased from 13 to 35°C, whereas the amount of radon adsorbed decreased by a factor of 3 when the relative humidities were increased from 15 to 90% at 35°C. Also, Sextro and Lee (*17*) studied the performance of both the open-faced and the diffusion-limited charcoal canisters at varying radon concentrations. The errors ranged from 10 to 105% for the open-faced canisters and from 77 to 115% for the diffusion-limited canisters. The data were taken by using from two to six canisters of each type. The discrepancies in the above studies demonstrate that the effects of temperature and humidity on radon adsorption are not clearly understood.

The objective of the present research was to study the adsorption of radon on activated carbon in the presence of water vapor under static conditions. The equilibrium data were obtained by simultaneously measuring gas- and solid-phase radon concentrations by the use of a specially designed adsorption apparatus.

EXPERIMENTAL APPARATUS AND PROCEDURE

Material

A coal-based activated carbon, Type BPL, was provided by Calgon Carbon Corporation for use in this study. It had a specific surface area of 874 m^2/g with an average pore diameter of 26.1 Å. The pore volume for the 6 × 16 mesh particles was 0.57 cm^3/g. Radon gas was generated by flowing dry nitrogen through a Model Rn-1025 Pylon source developed by Pylon Electronics Development Ltd., Canada. The source contained a dry powder of ^{226}Ra which had a stated activity of 22.6 kBq. The source was capable of producing 76.87 pCi/L of radon gas.

Apparatus and Procedure

The uptake of water by the activated carbon was measured with a Cahn-D100 electrobalance which was used to measure the weight of water adsorbed as a function of pressure. The apparatus was modified so that radon adsorption could be measured simultaneously in the presence of the water vapor. The concentration of radon in the gas phase was measured by using a Lucas Cell along with an AB-5 radiation monitor. The gas-phase count was based on the decay of alpha-emitting radon daughters (^{218}Po and ^{214}Po). The radon concentration in the solid phase was determined simultaneously by measuring the gamma activity with an Na(Tl) detector. A count time of at least 10 minutes was required to obtain a statistically significant count. A detailed description of the apparatus is given by Hassan et al. (*8*).

Approximately 5 g activated carbon stacked in 2 or 3 layers on the bottom of the sample tube were used in all experimental runs. When larger amounts were used, very long times were required before any significant count could be obtained from the detector. When smaller quantities were used, counts from the detector were insignificant, relative to the background level, even after adsorption was continued for 40 hours.

Each activated carbon sample was regenerated by heating it under a vacuum at a temperature of 573 ± 0.1 K for 12 hours. After regeneration, the sample was cooled to the desired adsorption temperature, and a background count was obtained from the Lucas cell. The calibration method used prior to each run and the radioactivity calculation are described in a previous paper [Hassan et al. (*8*)]. The sample-holding tube containing the carbon was placed directly on the top of the NaI(Tl) detector, and moist radon-laden nitrogen was introduced into the system in small pressure increments of approximately 50 mmHg. After each incremental pressure increase, 3.5 hours were allowed for radon and its daughters that had been adsorbed on the solid to reach radioactive equilibrium. It should be noted that radioactive equilibrium is different from physical adsorption equilib-

rium which is established between the radon in the gas and on the adsorbent in approximately 15 minutes (*8*). Once radioactive equilibrium was reached, the gas-phase and solid-phase counts were obtained simultaneously. Subsequent data points were obtained by admitting more radon-laden nitrogen into the system and following the same procedure.

Binary mixtures of water and radon in nitrogen were prepared by bubbling the dry radon-laden nitrogen through water contained in two saturators. The two saturators, which were arranged in series, were immersed in a constant temperature bath whose temperature was controlled within ±0.1 K. The desired relative humidity of the gas stream was maintained by varying the temperature of the bath. The radon concentration of the nitrogen stream was continuously monitored by the Lucas cell to ensure that the water first became saturated with radon. When the outlet radon concentration became equal to the inlet concentration, and the gas stream became humidified with water vapor to the desired relative humidity level, the mixture was collected in a 2.8-L glass surge chamber prior to introducing it into the adsorption system. In order to have an adequate amount of radon and water vapor to completely load the adsorbent at any temperature, the 2.8-L surge chamber was continuously replenished with radon and water vapor in nitrogen at the desired concentration levels. The continuous replenishment also eliminated the build-up of radon daughters in the surge chamber. As noted earlier, the amount of water adsorbed was determined by measuring its uptake with the electrobalance. The final water concentration in the vapor phase was determined by measuring the pressure change in the system and from the known volume of the adsorption system.

RESULTS AND DISCUSSION

Pure component equilibrium data of radon and water vapor are shown in Fig. 1. The isotherm for water vapor was of Type V whereas it was Type III for radon. Because the surface of activated carbon is generally nonpolar in nature, the uptake capacity of water vapor at low relative humidities is extremely small. As adsorption progresses, the adsorbed water can promote further adsorption through hydrogen bonding. However, water condenses in the pores at higher relative pressures, which results in a sharp rise in the uptake. Type III isotherms are characteristics of systems in which adsorbent–adsorbate interactions are very weak. Although radon is a radioactive gas, it is still inert. Therefore, radon might have been expected to give a Type III isotherm. The isotherm data of Burtt and Kurbatov (*1*), Coleman et al. (*4*), and Przytycka (*12*) also exhibited Type III characteristics.

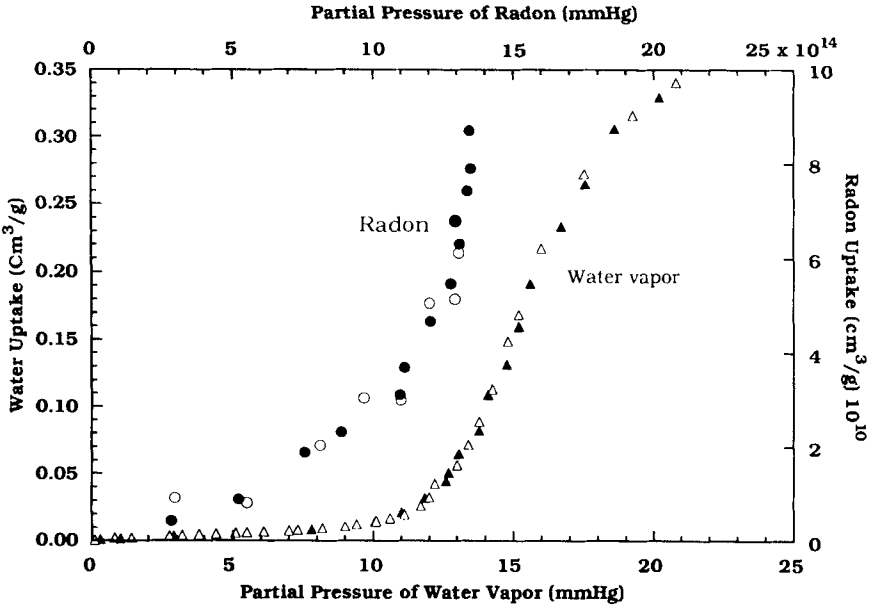

FIG. 1. Adsorption isotherms of pure water vapor and radon on BPL activated carbon at 298 K.

The error in the count rate of the gas phase was determined at the one sigma significance level and it ranged from 4.79 to 11.92%. The minimum detectable activity due to background count was found to be 21.2 counts per minute, which is eqivalent to 5.0×10^{-12} cm^3 of radon per minute at the three sigma significance level. Adsorption measurements were repeated for each run to check the reproducibility of the experimental data; the data were reproducible, with an average error of less than 5%.

The equilibrium data of radon–water vapor mixtures were obtained by using two approaches. First, activated carbon was preequilibrated with water vapor at a desired relative humidity, i.e., 40, 60, or 80%. The radon-laden dry nitrogen was then introduced into the system in small pressure increments of approximately 50 mmHg, and radon and its daughters were allowed to equilibrate in the usual manner. The uptake of radon on the carbon that had been preequilibrated with water was measured at 288 and 298 K and is shown in Figs. 2 and 3, respectively. At 303 K, the uptake of radon was very small and the gamma count was statistically insignificant. The isotherm data for radon at both temperatures were Type III. The amount of radon adsorbed on activated carbon that had been preequilibrated with water was lower than the amount adsorbed on dry carbon.

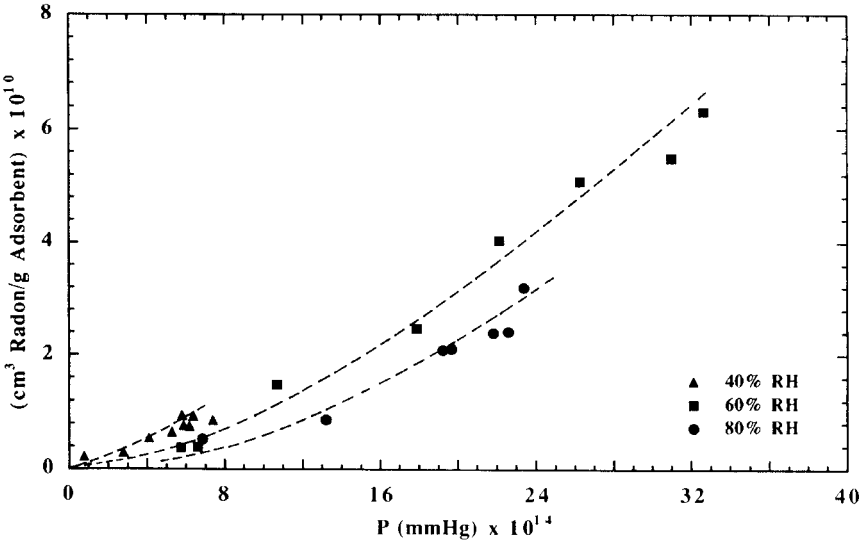

FIG. 2. Radon adsorption isotherms on preequilibrated BPL activated carbon at 288 K.

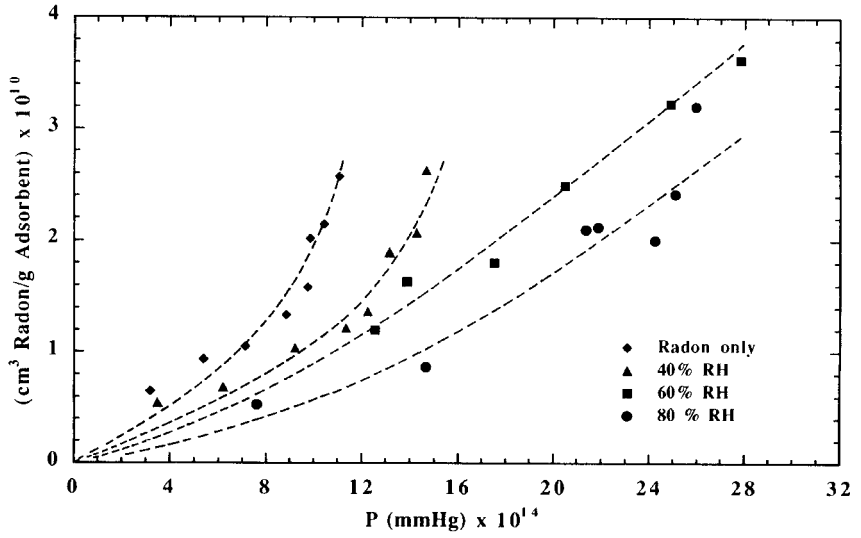

FIG. 3. Radon adsorption isotherms on preequilibrated BPL activated carbon at 298 K.

Also, the adsorption capacity decreased with an increase of both temperature and relative humidity. It is interesting to note that when radon-laden nitrogen was introduced into the system, a small amount of water vapor was desorbed from the activated carbon. In a separate run, radon-free nitrogen was introduced into the system under similar conditions and the same results were observed, suggesting that either nitrogen or some impurity in nitrogen was displacing water vapor from the carbon surface. The interaction between radon and the carbon does not appear to be strong enough to displace the preadsorbed water vapor. Thomas (*19*) and Strong and Levins (*18*) made the same conclusion from the results of their dynamic adsorption studies.

In the second approach, mixtures of radon and water vapor were directly admitted into the dry activated carbon. The adsorption capacities at 298 K are compared in Fig. 4 with the data obtained on the carbon that had been preequilibrated with water. The shape of the isotherms of radon from the radon–water vapor mixture was also of Type III. A similar trend (a decrease in the adsorption capacity with an increase in relative humidity) was observed. The uptake of radon was significantly lower than the amount adsorbed from the dry nitrogen. The adsorption capacity for radon was

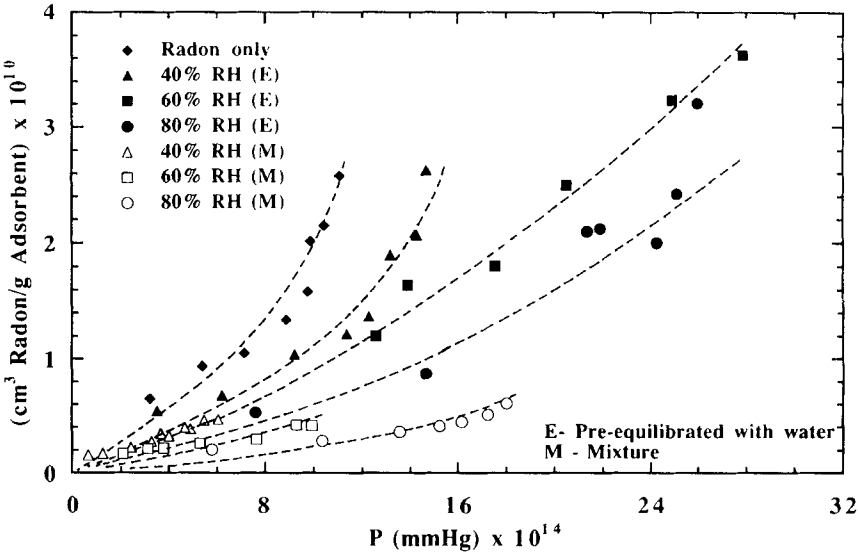

FIG. 4. Comparison of radon uptake from water vapor mixture with that on preequilibrated BPL activated carbon at 298 K.

also lower when radon was adsorbed from the radon–water vapor mixtures than when the carbon was first preequilibrated with water vapor. This can be attributed to the stronger adsorption of water molecules.

When the radon–water vapor mixture was introduced, the temperature increased slightly due to the exothermic heat of adsorption of the water vapor, which possibly results in a reduced radon uptake. A similar observation was reported by Thomas (*19*), who investigated the adsorption of radon on activated carbon in a dynamic system. According to Thomas, a rise in temperature of about 7°C, which he attributed to the heat of adsorption of water, was responsible for the decrease in uptake capacity of radon by the carbon. However, the greater uptake capacity of radon by the carbon that was preequilibrated with water may be attributed to the absorption of radon in the water that was contained in the micropores of the adsorbent. It should be noted that the solubility of radon in water is relatively high. The Henry's law constant for radon in water is 3.79×10^{14} pCi/(g-atm), whereas for radon on dry BPL activated carbon it is 1.67×10^{16} pCi/(g-atm). As a consequence, 44 times more radon will dissolve in the condensed water in the carbon than would have been adsorbed on the carbon surface. Since both radon adsorption and absorption are occurring when the carbon is preequilibrated with water vapor, the uptake capacity of radon on BPL activated carbon should be expected to be higher than when radon was adsorbed from the mixture. This is shown in Fig. 4. The amount of water adsorbed by activated carbon at a relative humidity of 40% is extremey small. Therefore, the absorption effect will be negligible; the small differences in the radon uptake can be attributed to the temperature difference. A sharp increase in the water uptake occurs around 50% relative humidity due to pore filling, with the amount of water adsorbed at 288 K being higher than that at 298 K, as expected. The large difference in the uptake of radon at the two temperatures as the relative humidity of the nitrogen stream was increased to 60 and 80% can be attributed to the absorption of radon in water. If a longer period of time is allowed, the amount adsorbed from the mixture should eventually be equal to the preequilibrated value. However, an experimental run typically took 3.5 days and no substantial difference was observed during that time period, suggesting that the diffusion of radon in the water inside the pores is very slow.

As noted earlier, there is strong disagreement in the ltierature on the effect of moisture on radon adsorption by activated carbon. Hassan et al. (*7*) compared the literature data for water vapor adsorption by the same type of activated carbon (Type BPL) and found that the uptake capacity can vary significantly. As a consequence, the radon uptake will also vary.

Correlation of Equilibrium Data

The Freundlich equation has been modified to correlate the equilibrium adsorption data of radon on activated carbon that had been preequilibrated with water vapor. The modified equation can be written as

$$q = k'\left(\frac{P}{P_0}\right)^n \tag{1}$$

or

$$\ln q = \ln k' + n \ln\left(\frac{P}{P_0}\right) \tag{2}$$

where q is the amount of gas adsorbed, P is the system pressure, and P_0 is a reference pressure and is arbitrarily set to 10^{-14} mmHg. Here k' is a measure of the volume of the gas adsorbed per unit mass of adsorbent, and n is the intensity of adsorption. The temperature dependence of k' was previously determined from a correlation of pure radon adsorption data with Eq. (1) (*8*) for a value of n equal to 1.75, and it can be expressed as

$$k'_{(T)} = 2.10 \times 10^{-11} - 6.58 \times 10^{-14}\, T \tag{3}$$

Thus, the amount of radon adsorbed on BPL activated carbon corresponding to a specific gas-phase concentration can be expressed as

$$q = (2.10 \times 10^{-11} - 6.58 \times 10^{-14}\, T)\left(\frac{P}{1 \times 10^{-14}}\right)^{1.75} \tag{4}$$

Knowing the solid-phase concentration, the gas-phase concentration can be obtained in picocuries per liter from the equation

$$C = \frac{5.48 \times 10^3\, q^{0.571} T^{-1}}{(2.10 \times 10^{-11} - 6.58 \times 10^{-14}\, T)^{0.571}} \tag{5}$$

The coadsorption data of radon and water vapor on BPL activated carbon at 298 K are correlated with a different version of Eq. (1) suggested by McGavack and Patrick (*9*):

$$q = k_1\left(k_2 \frac{P}{P_0}\right)^n \tag{6}$$

where parameters k_1 and k_2 are assumed to be a function of relative humidity only. The dependence of the parameters k_1 and k_2 on relative humidity at 298 K was obtained for a value of n equal to 1.75, and the results are shown in Figs. 5 and 6. The parameters can be expressed in terms of percent relative humidity (%RH) as

$$k_1 = 2.25 \times 10^{-11} - 1.49 \times 10^{-13} \times \%\text{RH} \tag{7}$$

and

$$k_2 = 2.77 + 2.01 \times 10^{-12}\ \%\text{RH} \tag{8}$$

The amount of radon adsorbed on BPL activated carbon at 298 K and at any relative humidity may then be expressed as

$$q = (2.25 \times 10^{-11} - 1.49 \times 10^{-13} \times \%\text{RH}) \times \left(\frac{2.77 + 2.01 \times 10^{-12} \times \%\text{RH}}{1.0 \times 10^{14}} P\right)^{1.75} \tag{9}$$

Figure 7 shows that the modified Freundlich equation describes radon adsorption data relatively well with an average error of ±8%. The symbols

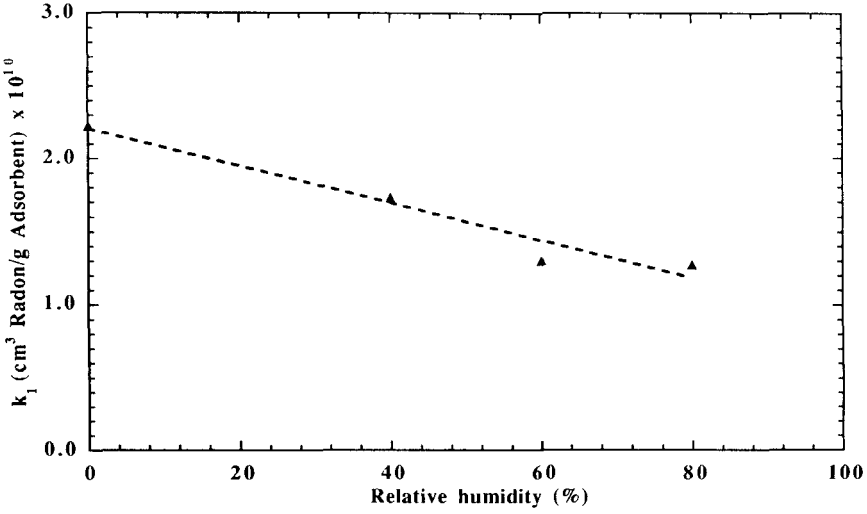

FIG. 5. Relative humidity dependence of the parameter k_1 in the modified Freundlich equation at 298 K.

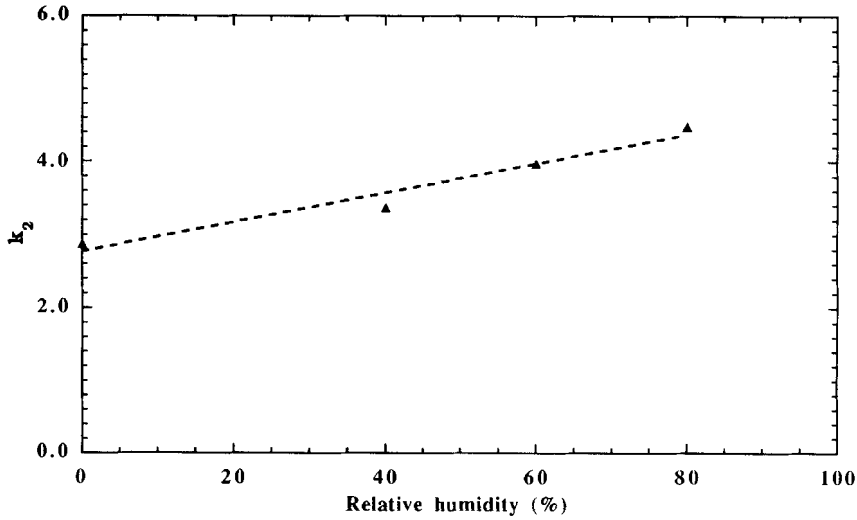

FIG. 6. Relative humidity dependence of the parameter k_2 in the modified Freundlich equation at 298 K.

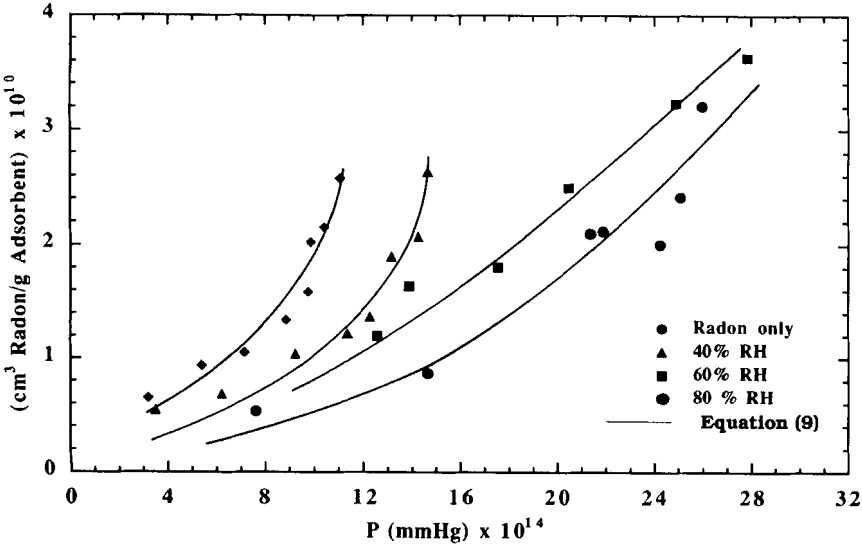

FIG. 7. Correlation of radon coadsorption data according to the modified Freundlich equation at 298 K.

in the figure are experimental data for different relative humidities, and the solid lines represent the predicted curves.

The equilibrium isotherms of radon, such as the ones shown above, can be used for measuring radon concentrations in homes, provided accurate isotherm data are available for the activated carbon. From knowledge of the solid-phase radon concentration, the equilibrium isotherm (Eq. 9) corresponding to the particular field conditions, temperature, and humidity can be used to determine the concentration of radon in the gas phase.

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