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Adsorption of Radon and Water Vapor on Commercial Activated Carbons

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ABSTRACT

Equilibrium adsorption isotherms are reported for radon and water vapor on two commercial activated carbons: coconut shell Type PCB and hardwood Type BD. The isotherms of the water vapor were measured gravimetrically at 298 K. The isotherms of radon from dry nitrogen were obtained at 293, 298, and 308 K while the data for the mixture of radon and water vapor were measured at 298 K. The concentrations of radon in the gas and solid phases were measured simultaneously, once the adsorption equilibrium and the radioactive equilibrium between the radon and its daughter products were established. The shape of the isotherms was of Type III for the radon and Type V for the water vapor, according to Brunauer's classification. The adsorption mechanism was similar for both the radon and the water vapor, being physical adsorption on the macropore surface area in the low pressure region and micropore filling near saturation pressure. The uptake capacity of radon decreased both with increasing temperature and relative humidity. The heat of adsorption data indicated that the PCB- and the BD-activated carbons provided a heterogeneous surface for radon adsorption. The equilibrium data for radon were correlated with a modified Freundlich equation.

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INTRODUCTION

There is an increasing concern about the potential health risk of radon and its daughter products in indoor air. In response to this, a considerable effort led by the Environmental Protection Agency, as well as numerous public agencies and private companies, has been directed to understand and alleviate the problem. Radon is a radioactive gas (half-life 3.82 days) produced by the decay of naturally occurring radium in soil and rock, and it can accumulate in certain homes and buildings to levels of health concern. Radon further decays into short-lived daughter products (^{218}Po, ^{214}Pb, ^{214}Bi, and ^{214}Po with half-lives 3.05 minutes, 26.8 minutes, 19.7 minutes, and 1.6×10^{-4} second, respectively) that emit α-particles, and γ- and β-radiation. The α-particles can be directly inhaled or ingested after attaching to airborne particles. Upon inhalation, they can lodge in the lungs, where they can ultimately cause tissue damage and cancer.

Several different methods are available for measuring radon concentrations in homes. One method that is most frequently used and currently recommended by the EPA utilizes radon adsorption on activated carbon and subsequent counting of the γ-radiation emitted by ^{214}Pb and ^{214}Bi radon daughters. Rutherford (1) was the first to recognize that radon in air can be adsorbed on activated carbon, but only recently has it been employed for designing radon detectors, better known as charcoal canisters, which are widely used to measure the concentration of radon in indoor air (2–6). The design of the charcoal canisters was based on the assumption that the equilibrium relationship between the gas phase and measured solid phase radon concentration is linear, with a slope equal to 1. A concise discussion of the theory and underlying assumptions of radon adsorption on activated carbon in canisters was given by Cohen and Cohen (2).

Adsorption isotherm data of radon on activated carbon were reported in the literature between 1940 and 1960. The data of Gubeli and Stammbach (7), which are in the form of adsorption isotherms of radon in air on $ZnCl_2$-activated carbon, was taken in the temperature range 273–393 K. Later, Gubeli and Stori (8) used elm-wood carbon for the adsorption of radon in the presence of various gases at atmospheric pressure. The equilibrium isotherms of radon on both elm-wood and $ZnCl_2$-activated carbons were linear. Coleman et al. (9) measured the adsorption isotherms of radon on coconut shell carbon at 25°C and radon partial pressures in the range 1.4×10^{-15} to 6.0×10^{-14} atm. The data of Coleman et al. exhibited a considerable deviation from linearity when the amount of radon adsorbed was plotted as a function of its partial pressure in air. The equilibrium isotherms measured by Przytycka (10) for radon adsorption on three types

of activated carbons at 20°C also exhibited a nonlinear relationship for the amount of radon adsorbed with pressure. Recently, Hassan et al. (11) obtained the adsorption isotherms of radon from dry nitrogen on BPL-activated carbon at 288, 298, and 308 K using a special experimental apparatus designed for simultaneous measurement of radon concentration in the gas and solid phases. This new system was presumably far more efficient than conventional ionization chambers, which are well-known for the difficulties involved in measuring small ionization currents. Later, Hassan et al. (12) reported on adsorption of radon from a humid atmosphere on BPL-activated carbon and found that the water vapor present in the mixture reduced the uptake capacity for radon due to strong competition by water vapor molecules for available sites on the carbon. Water is present in humid, radon-laden air, and during field measurements, water is coadsorbed on activated carbon in canisters. Proper design of the canisters requires a comprehensive understanding of how water affects the adsorption of radon on the carbon by measuring equilibrium isotherm data for pure components and for mixtures of radon and water vapor. At present, these equilibrium isotherm data are limited, particularly for commercial activated carbons.

The objective of this study was to obtain equilibrium isotherm data of radon from dry nitrogen and in the presence of water vapor on two commercially available activated carbons: coconut shell (Type PCB) and hardwood (Type BD) activated carbons. The radon concentrations in the gas and solid phases were measured simultaneously under static conditions. The isotherms of radon from dry nitrogen were measured at three different temperatures whereas the isotherms of radon in the presence of water vapor were obtained at three prescribed levels of relative humidity. The isotherms for adsorption of pure water vapor on the activated carbons were also measured gravimetrically at room temperature. The equilibrium isotherm data of radon were correlated according to a modified Freundlich equation. The isosteric heats of adsorption were calculated from experimental isotherm data at different adsorbent loading.

EXPERIMENTAL SECTION

Materials

The activated carbons used in this study were coconut shell (Type PCB) and hardwood (Type BD) carbons provided by Calgon Carbon Corporation. The properties of the carbons, along with coal-based activated carbon (Type BPL), are given in Table 1. Radon gas was generated from a Model Rn-1025 Pylon flow-through source and was made available by flowing dry nitrogen gas from a cylinder. The source was a sealed, dry

TABLE 1
Physicochemical Properties of Activated Carbons

Property	Type BPL	Type PCB	Type BD
Particle size (Å)[b]	6 × 16 mesh	6 × 12 mesh	8 × 12 mesh
Surface area, S (m^2/g):[a]			
Micropores	823	663	294
Meso and macropores	9	50	101
Total	832	713	395
Pore volume, V (cm^3/g):[a]			
Micropores	0.47	0.38	0.14
Meso and macropores	0.10	0.20	0.27
Total	0.57	0.58	0.41
Average pore diameter, $4V/S$ (Å)[a]	26	24	41.7[c]
Bulk density (g/cm^3)[b]	0.60	0.72	0.72
Equilibrium water capacity (% by wt)[b]	—	—	29.5
Moisture content as shipped (% by wt)[b]	<1	—	<1.5

[a] Analysis made by Porous Materials, Inc., Ithaca, New York.
[b] Analysis provided by the manufacturer.
[c] Based on total surface area and pore volume.

powder containing ^{226}Ra with a stated activity of 22.6 kBq. The source was capable of producing a constant radon gas at a rate of 2.847 Bq/min (76.87 pCi/min.). A certified γ-source, Model Ra-226-Sc, was obtained from The Nucleus, Inc., Oak Ridge, Tennessee. The source, with a stated activity of 548 Bq (14,800 pCi), was used as a reference to calibrate the NaI(TI) spectroscopy system.

Experimental Apparatus and Procedure

The adsorption isotherm data of radon and water vapor were measured using the experimental system shown in Fig. 1. This system consists of a Cahn D-100 electrobalance connected to an all-glass apparatus that has been designed to measure radon concentration simultaneously in the gas and solid phases. The sample holding tube was a 48 mm diameter × 500 mm long glass tube that was made with a flat bottom to match the geometry of the γ-ray calibration source. The bottom of the tube was positioned to provide direct contact with a lead-shielded 50 mm × 50 mm NaI(TI) detector, coupled to a multichannel pulse height analyzer and associated electrons. An α-radiation monitor, Pylon AB-5 along with Lucas cell, was connected on-line to the vacuum bottle housing the electrobalance, a gas reservoir, a Model Rn-1025 Pylon flow-through source, and a vacuum pumping arrangement. The entire system could be evacuated to a pressure

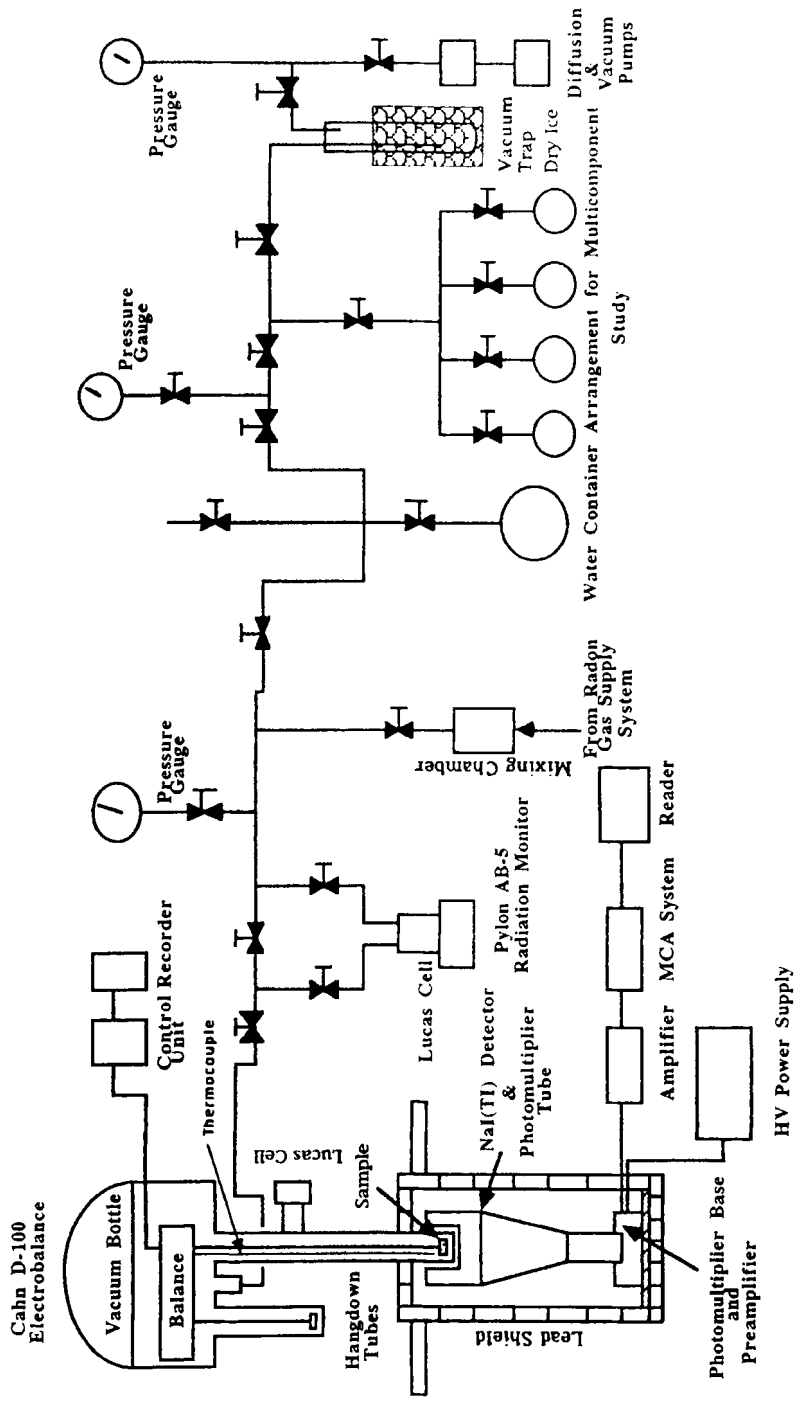

FIG. 1 Schematic flow diagram of radon adsorption apparatus.

of 10^{-4} mmH by a Model 1400 Sargent-Welch roughing pump, a carbon dioxide cold trap, and two type VMF diffusion pumps. System pressures up to 10^{-3} mmHg were monitored with two covectron gauges, while pressures below 10^{-4} were measured with two ionization gauges; equilibrium pressures up to 800 mmHg were measured with Wallace and Tiernan absolute pressure gauges. A detailed description of the experimental apparatus, the methods of calibration of the detectors prior to each adsorption run, and the radioactivity calculation was given by Hassan et al. (11, 12).

The adsorbent sample was regenerated by heating it at 573 ± 0.1 K in a vacuum of 10^{-4} mmHg for 12 hours. After regeneration, the sample was cooled to the desired adsorption temperature, and a background count for the Lucas cell was obtained. The sample holding tube was then placed directly on the top of the NaI(Tl) detector, and radon-laden dry nitrogen was introduced into the system in small pressure increments of approximately 50 mmHg. After each increment, a period of 3.5 hours was allowed for radon and its daughter products to reach radioactive equilibrium; adsorption equilibrium between radon in the gas phase and that in the solid phase was usually complete in 15 minutes as previously verified (11). Once radioactive equilibrium was reached, 10-minute counts for γ-activity arising from ^{214}Pb and ^{214}Bi nuclei were recorded by the lead-shielded 50 mm × 50 mm NaI(Tl) detector, coupled to a multichannel pulse height analyzer and associated electronics. The counts were obtained in the region of interest, which comprised of the energy photopeaks 242, 295, and 395 keV for ^{214}Pb, and 609 keV for ^{214}Bi. The total counts below these photopeaks were corrected for background radiation that originated from radioisotopes present in the vicinity of the detector and natural radioactivity. The number of moles of radon adsorbed per unit weight of adsorbent was then obtained from the total net count rate of the ^{214}Pb or ^{214}Bi energy peaks, the efficiency of the energy peaks that was determined from calibration of the NaI(Tl) detector, and the probability of emission of the γ-rays for each disintegration of the ^{214}Pb or ^{214}Bi nuclei using the relation given by Mann et al. (13). The volume of radon in the solid phase was calculated from the number of moles of radon per unit weight, the atomic weight of radon, Avogadro's number (6.02×10^{23} atoms/mol), and the ideal gas law.

The concentration of radon in the gas phase was obtained by measuring the counts due to α-particles produced by the decay of radon and its daughters (^{218}Po and ^{214}Po). Counting in the gas phase was performed using the flow-through Lucas cell, along with the AB-5 Radiation Monitor provided by Pylon Electronics Co. Ottawa, Canada. The number of moles of radon in the gas phase was calculated from the net count rate of the α-particles, the counting efficiency of the Lucas cell that was determined

during calibration, the volume of the Lucas cell, and the decay factor for radon from the midpoint of the time when radon was admitted into the system until counting was started. The ideal gas law was used to calculate the partial pressure of radon.

RESULTS AND DISCUSSION

Adsorption Isotherms for Water Vapor

The isotherms of pure water vapor on BPL-, PCB-, and BD-activated carbons were measured gravimetrically at 298 K by using a Cahn D-100 electrobalance and a standard adsorption procedure. The results are shown in Fig. 2, where the open and solid symbols represent two isotherms obtained at the same temperature. The data were reproducible with an average error of less than 1%. It can be seen that the shape of the isotherms of water was of Type V, according to Brunauer's classification (14). These isotherms show a small uptake of water in the low pressure region, followed by a rapid adsorption increase in the medium to saturation pressure. Because activated carbons generally differ in their structural

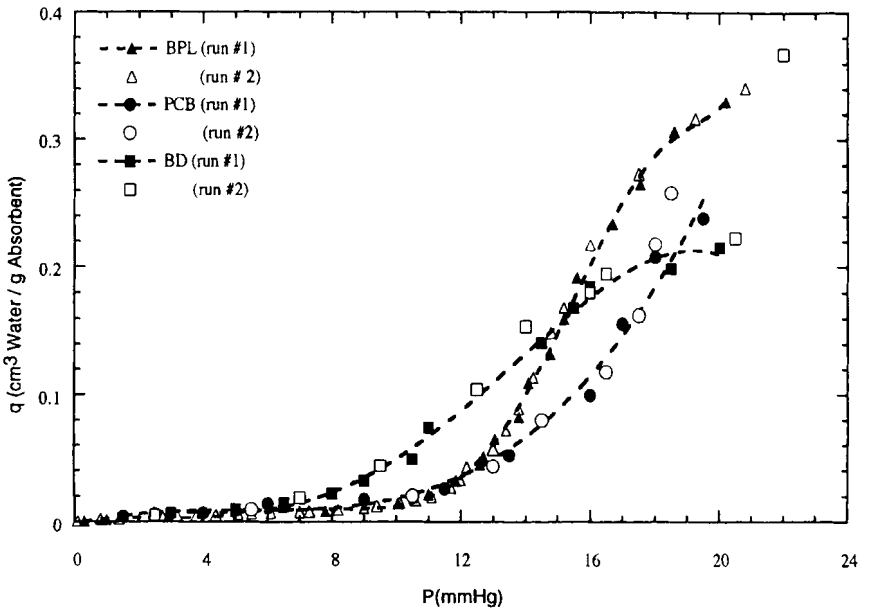

FIG. 2 Adsorption isotherms of water vapor at 298 K.

defects as well as in their surface area and pore size distribution, their uptake capacities vary. This is illustrated in Fig. 2, where the BD-activated carbon has a greater uptake capacity in the low pressure region than the BPL- and the PCB-activated carbons. In the intermediate to saturation pressure, the isotherms cross each other and the BPL-activated carbon exhibits a greater saturation capacity. This experimental finding is consistent with the Zsigmondy concept (15), which assumes that physical adsorption of the vapor occurs on the macropores surface area until capillary condensation commences in the micropores. In this case, the BD-activated carbon had approximately 25% of its total surface area as macropores, which were entirely available for physical adsorption; the BPL- and the PCB-activated carbons had 1 and 7% of their total surface areas as macropores, respectively. Because of the small macropore surface area of these two carbons, their uptake capacities were extremely small in the low pressure region. The uptake of water on the carbons then increased rapidly due to cooperative adsorption between water molecules through hydrogen bonding, followed by condensation in small pores (micropore filling). Thus, the BPL-activated carbon, which had 823 m^2/g of micropore surface area (99% of its total), shows the greatest saturation capacity, followed by the PCB-activated carbon with 663 m^2/g of micropore surface. The BD-activated carbon, which had only 294 m^2/g of micropore surface area (less than 75% of its total), shows a lower saturation capacity than the other two carbons.

Adsorption Isotherms for Radon

The isotherms of radon from dry nitrogen on PCB- and BD-activated carbons were obtained at three temperatures: 293, 298, and 303 K. The equilibrium data in radioactivity units are given in Tables 2 and 3. The error associated with the count rate of the gas phase was determined at the one sigma significance level and it ranged from approximately 5 to 12%. The minimum detectable activity due to background count was found to be about 21 counts per minute at the three sigma significance level. The equilibrium data in SI units are presented in Figs. 2 and 3. The shape of isotherms for radon was of Type III (in Brunauer's classification). This is typical of systems in which adsorbate–adsorbent interaction is relatively weak. The two carbons generally exhibited similar behavior, i.e., increasing adsorption capacity at decreasing temperature. The isotherms of radon on the PCB- and the BD-activated carbons are compared in Fig. 4 with that of the BPL-activated carbon at 298 K. The adsorption of radon was greater for the BD-activated carbon than for the BPL- and the PCB-activated carbons. The large difference in the uptake values can be attributed

TABLE 2
Equilibrium Adsorption Data of Radon on PCB-Activated Carbon

293 K		298 K		308 K	
pCi[a] of Rn/L of N_2	pCi of Rn/g of carbon	pCi of Rn/L of N_2	pCi of Rn/g of carbon	pCi of Rn/L of N_2	pCi of Rn/g of carbon
14.0	71.6	17.8	86.7	25.6	72.8
25.5	102.8	50.9	144.4	36.0	93.1
40.2	123.2	62.4	161.1	42.6	100.0
45.3	171.0	73.6	173.3	46.3	111.8
56.6	108.9	80.5	234.7	51.4	122.5
66.1	209.3	91.4	227.6	62.0	129.7
68.7	216.5				

[a] A picocurie is an amount of radioactive material corresponding to 2.22 disintegrations per minute.

to the differences in the surface area as well as the broad pore size distribution. This finding is again consistent with the results for water vapor adsorption on the carbons despite the difference in the shape of the isotherms of the radon and the water vapor on the carbons. It can be concluded from both experimental data that physical adsorption on the macropore surface area is important in the low pressure region whereas the micropore filling by condensation controls the adsorption mechanism in the high pressure region.

The equilibrium data of radon in the presence of water vapor can be obtained by using one of two approaches. In one approach the activated carbon is preequilibrated with water vapor at a desired relative humidity

TABLE 3
Equilibrium Adsorption Data of Radon on BD-Activated Carbon

293 K		298 K		308 K	
pCi of Rn/L of N_2	pCi of Rn/g of carbon	pCi of Rn/L of N_2	pCi of Rn/g of carbon	pCi of Rn/L of N_2	pCi of Rn/g of carbon
12.3	102.7	13.8	101.1	12.9	74.6
20.8	137.5	21.6	125.8	22.6	88.5
31.1	177.1	25.7	135.9	34.0	127.3
37.2	189.0	31.6	144.0	47.9	148.5
39.2	197.6	40.3	166.4	60.0	166.6
54.0	200.4	46.3	196.0	76.8	206.1

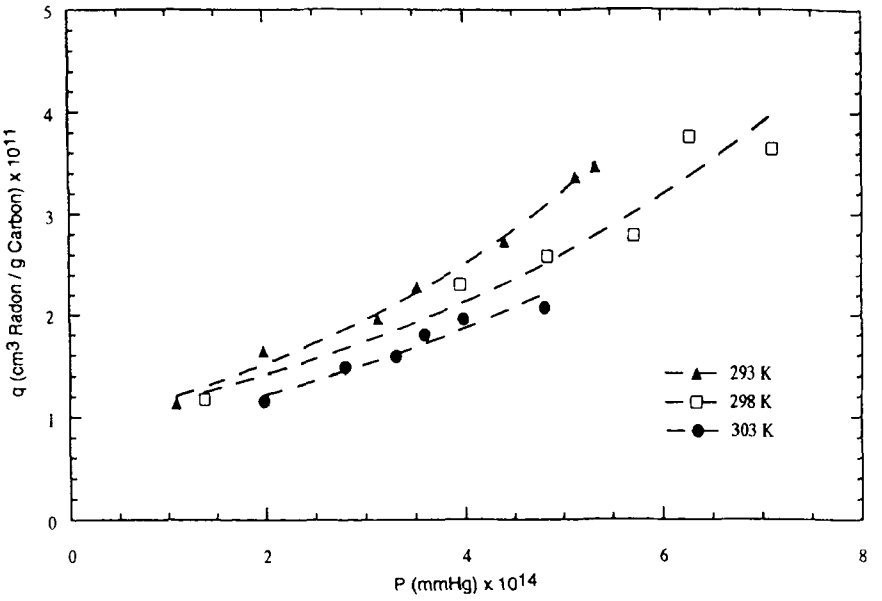

FIG. 3 Adsorption isotherms of radon from dry nitrogen on PCB activated carbon.

and the radon-laden dry nitrogen is then introduced into the system in small pressure increments. In the second approach a mixture of radon and water vapor in nitrogen is prepared and directly admitted into the dry activated carbon. Hassan et al. (12) compared these two approaches for radon adsorption on BPL-activated carbon and found that the uptake capacity was lower when radon was adsorbed from the mixture on dry activated carbon. Because the solubility of radon in water is relatively high, 44 times more radon was dissolved in adsorbed water in the preequilibrated carbon. In this study the isotherms of radon and water vapor mixture in nitrogen were measured at 298 K (Fig. 5), and the equilibrium data in radioactivity units are given in Tables 4 and 5. The uptake of radon from water vapor mixture is compared in Fig. 6 with that obtained when the PCB-activated carbon was preequilibrated with 40% relative humidity. It should be noted that the adsorption capacity was substantially lower when radon was adsorbed from the mixture than when the carbon was preequilibrated with water vapor. The adsorption isotherms of radon and water vapor mixture on the BD-activated carbon are shown in Fig. 7. The shape of the isotherms for radon in the presence of water vapor was also of

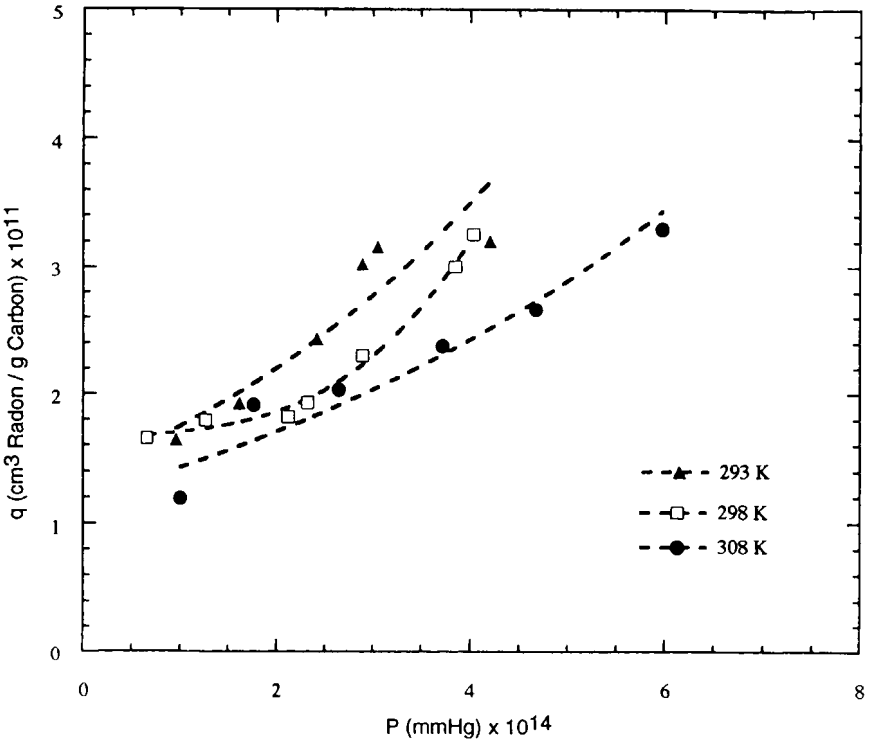

FIG. 4 Adsorption isotherms of radon from dry nitrogen on BD activated carbon.

Type III. The uptake of radon generally decreased with an increase in relative humidity. However, a sharp increase in radon uptake occurred when the relative humidity of the gas stream was increased from 60 to 80% and the temperature increased considerably.

Correlation of Equilibrium Data

The equilibrium adsorption data of radon from dry nitrogen were correlated according to a modified Freundlich equation, which is expressed in the form

$$q = k_1(P/P_0)^n \tag{1}$$

where q is the amount of gas adsorbed, P_0 is a reference pressure, k_1 is a measure of the volume of gas adsorbed per unit mass of adsorbent, and

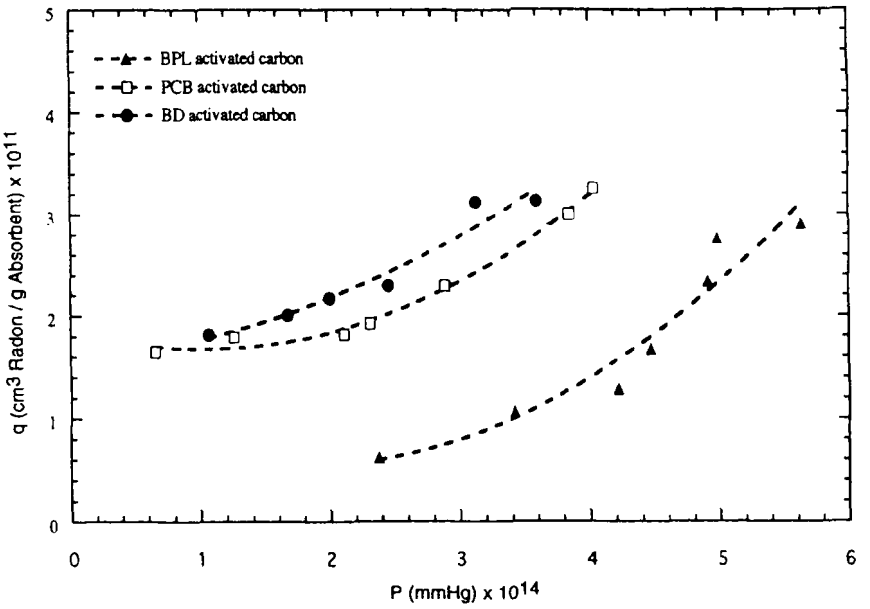

FIG. 5 Comparison of radon adsorption isotherms at 298 K.

TABLE 4
Equilibrium Adsorption Data of Radon–Water Vapor Mixture on PCB-Activated Carbon at 298 K

40% Relative humidity		60% Relative humidity		80% Relative humidity	
pCi of Rn/L of N_2	pCi of Rn/g of carbon	pCi of Rn/L of N_2	pCi of Rn/g of carbon	pCi of Rn/L of N_2	pCi of Rn/g of carbon
19.7	58.6	32.5	65.7	12.3	60.9
40.2	89.5	54.5	74.8	25.8	61.3
52.4	92.25	74.2	75.74	39.6	85.3
58.3	76.06	94.9	122.7	46.3	77.2
60.3	103.6	101.6	149.4	60.9	107.5
74	123.7	115	174.9	65.8	126
115.8	136	153.6	208.6	79.9	186.7

TABLE 5
Equilibrium Adsorption Data of Radon–Water Vapor Mixture on BD-Activated Carbon at 298 K

20% relative humidity		40% relative humidity		60% relative humidity	
pCi of Rn/L of N_2	pCi of Rn/g of carbon	pCi of Rn/L of N_2	pCi of Rn/g of carbon	pCi of Rn/L of N_2	pCi of Rn/g of carbon
12.3	60.9	19.7	58.6	32.5	65.7
25	61.3	40.2	90	54.5	74.8
39.6	85.3	52.4	92.3	74.2	75.7
46.3	77.2	58.3	76.1	94.9	123
60.9	107.5	60.3	104	102	149
65.8	126.1	74	124	115	175
79.9	186.7	116	136	154	209

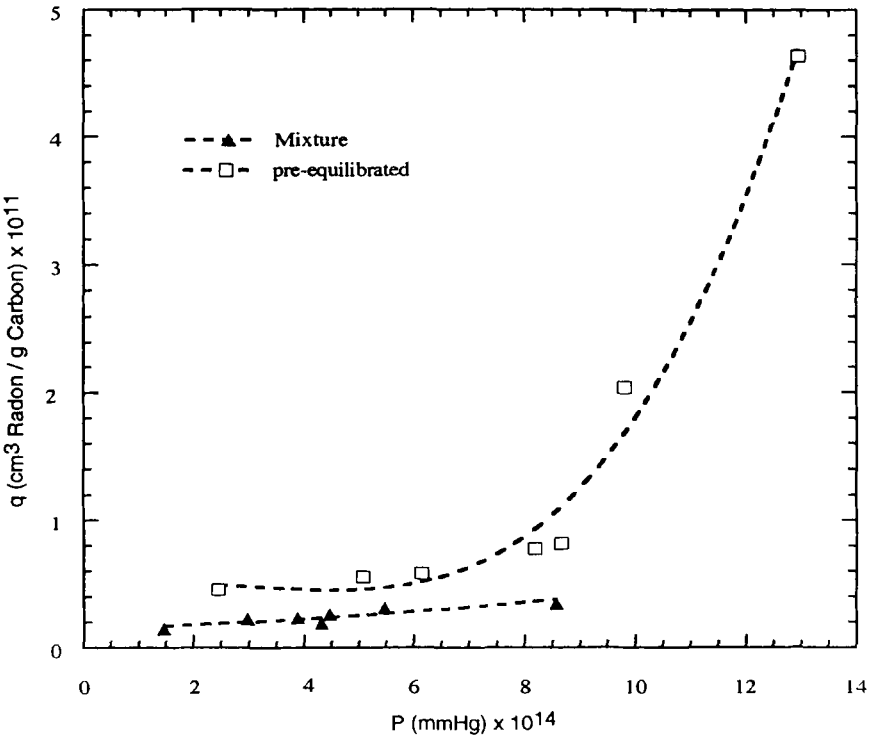

FIG. 6 Comparison of uptake values for radon from radon–water vapor mixture and on pre-equilibrated PCB activated carbon at 40% relative humidity and 298 K.

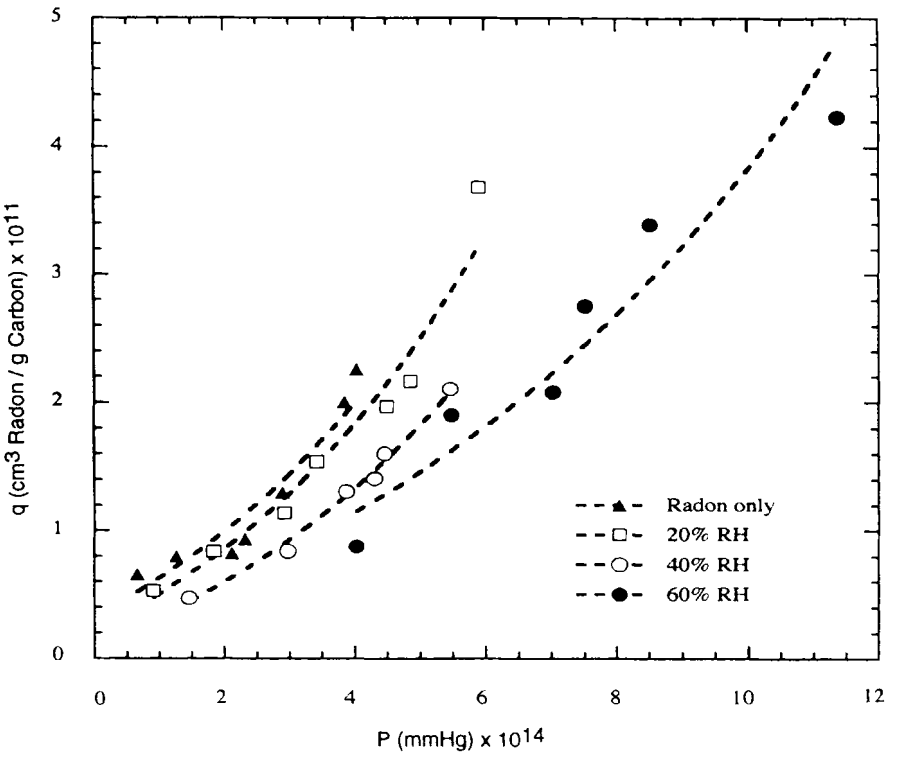

FIG. 7 Adsorption isotherms of radon–water vapor mixture on BD activated carbon at 298 K.

n is the intensity of adsorption. The reference pressure, P_0, was chosen arbitrarily by Hassan et al. (11) and was set at 10^{-14} to maintain the same order of magnitude as the system pressure. The reference pressure was allowed to vary in this correlation. A good fit of the experimental data and the values predicted by the modified Freundlich equation, with a value of n equal to 1.7 ± 0.3, was obtained as shown in Figs. 8 and 9. The values of parameters k_1 and P_0, along with the absolute average percent errors, are shown in Table 6. The maximum average error between experimental data and the theoretical values was approximately 7.5% for the PCB-activated carbon and 4.7% for the BD-activated carbon. The maximum errors were observed at 293 K. The temperature dependence of the parameter k_1 was found to be linear as shown in Fig. 9. It should be noted that the modified Freundlich equation is an empirical equation which is

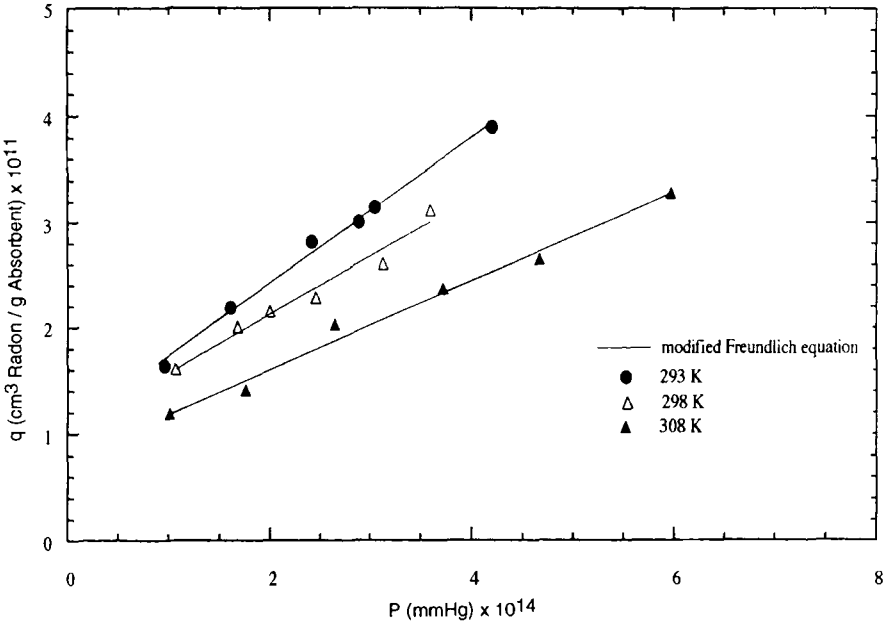

FIG. 8 Correlation of radon adsorption data for BD activated carbon according to the modified Freundlich equation (Eq. 1).

limited in its usefulness to its ability to fit the experimental data at one particular temperature. However, knowing the temperature dependence of the parameter k_1, the data can be interpolated to other temperatures.

Heat of Adsorption for Radon

The isosteric heat of adsorption of radon from dry nitrogen on PCB- and BD-activated carbons were obtained from the relationship given by Hersh (16):

$$H^{iso} = R[\partial \ln P/\partial(1/T)]q \quad (2)$$

where H^{iso} is the isosteric heat of adsorption, q is the amount of radon adsorbed per unit weight of adsorbent, P is the system pressure at equilibrium, R is the gas constant, and T is the adsorption temperature. The values of the heat of adsorption were calculated from the slope of the plot ln P versus $1/T$ at constant adsorbent loading. The variation of the heat of adsorption with adsorbent loading for the PCB- and the BD-activated

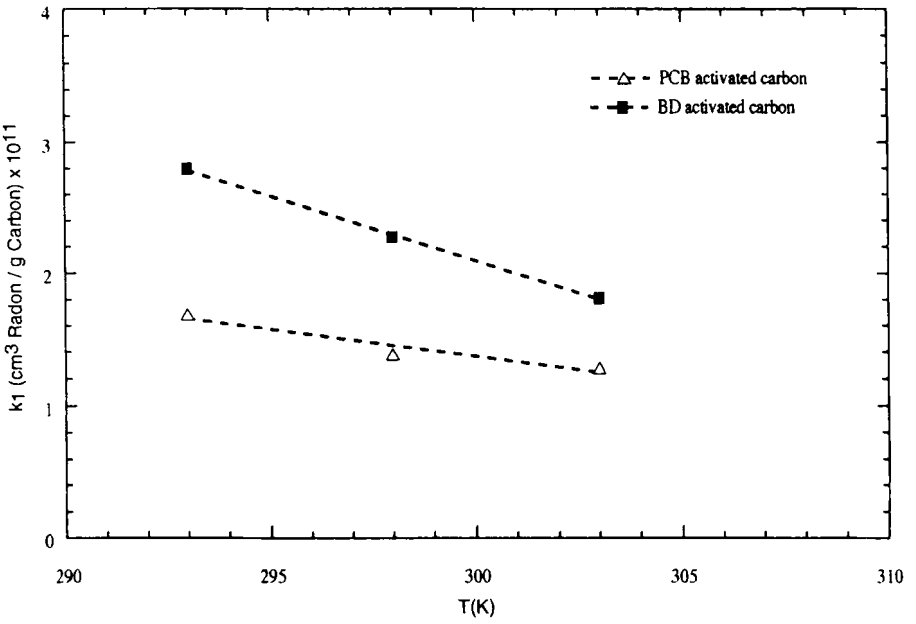

FIG. 9 Temperature dependence of the parameter k_1.

TABLE 6
Best Fit Parameters of the Modified Freundlich Equation

Temperature (K)	k_1 (cm^3 Rn/g carbon)	P_0 (mmHg)	Absolute average % error
	PCB-Activated Carbon		
293	1.69×10^{-11}	2.24×10^{-14}	7.47
298	1.38×10^{-11}	2.00×10^{-14}	7.10
303	1.28×10^{-11}	2.26×10^{-14}	2.53
	BD-Activated Carbon		
293	2.79×10^{-11}	2.44×10^{-14}	4.65
298	2.28×10^{-11}	2.19×10^{-14}	3.39
308	1.81×10^{-11}	2.33×10^{-14}	1.36

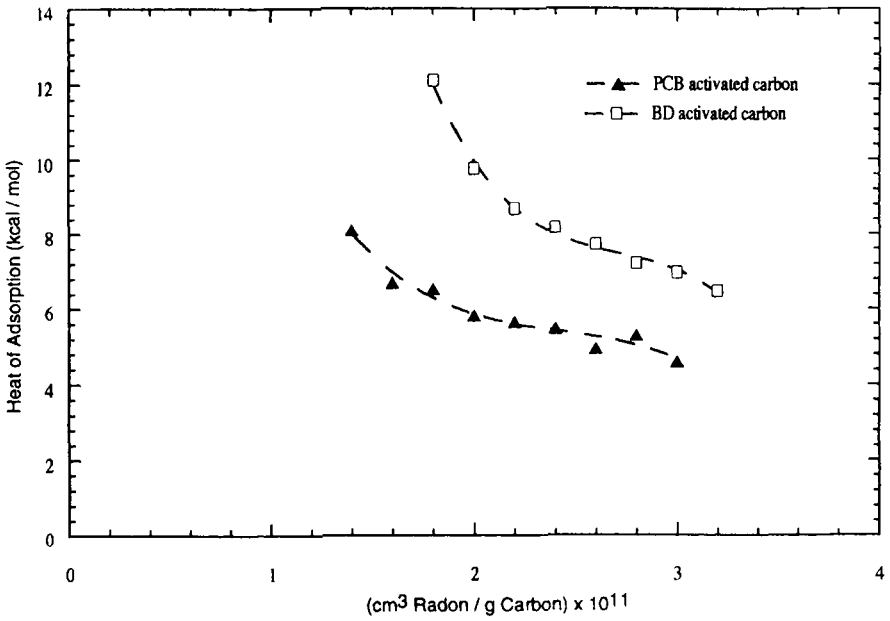

FIG. 10 Isosteric heats of adsorption for radon on activated carbons.

carbons is given in Fig. 10. It can be seen that the values for the heat of adsorption initially decrease with loading for both carbons and then gradually approach the latent heat of condensation for radon, which is approximately 4.01 kcal/mol. The decrease in the initial period of adsorption is typical for adsorption on heterogeneous surfaces. The isosteric heat of adsorption, which is a measure of the interactions between adsorbate molecules and the adsorbent lattice atoms, provides a valuable source of information on energetic heterogeneity of the surface.

NOMENCLATURE

k_1	a constant in the modified Freundlich equation
n	a constant in the modified Freundlich equation
P	equilibrium partial pressure of radon (mmHg)
P_0	reference pressure (mmHg)
q	volume of radon adsorbed per unit weight (cm^3 Rn/g adsorbent)
H^{iso}	isosteric heat of adsorption (kcal/mol)

R gas constant
T temperature (K)

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