

**Figure 6.** Plot of  $\ln W$  vs.  $\epsilon^m$  for the adsorption of water vapor at all temperatures on Mobil Sorbead R silica gel.

as required by the Dubinin-Polanyi theory (3).

The equilibrium data obtained in this study are compared in Figure 5 with the data presented by Mobil (15). As shown, the adsorption capacities presented herein are approximately 20% lower than Mobil's values. Although the regeneration temperature was not specified in that study, it may be concluded that the reactivation temperature must have been less than the value used in this work.

Bering et al. (16) showed that the volume of the adsorption space can be expressed by the relation

$$W = W_0 \exp[-(\epsilon/\beta E)]^m \quad (4)$$

where  $W_0$  is the volume of the adsorption space at saturation,  $E$  is a constant characteristic energy,  $\beta$  is the affinity factor, and  $m$  is a constant. Based on eq 4, a plot of  $\ln W$  vs.  $(\epsilon)^m$  should give a straight line with a slope of  $[-(1/\beta E)]^m$  and an intercept of  $\ln W_0$ . A value of  $m$  equal to 1.2 was found to give a straight line at all temperatures studied in this work (see Figure 6). The adequacy of the fit was checked by calculating the coefficient of determination; a value of 0.94 was found for this study. Huber et al. (17) reported that the values of  $m$  can be varied from 1.0 to 6.0 depending on the adsorbate-adsorbent system.

#### Glossary

$C$	BET constant
$E$	constant characteristic energy (kcal/g-mol)

$m$	constant in the generalized isotherm equation
$q_{st}$	isosteric heat of adsorption (kcal/g-mol)
$P$	equilibrium vapor pressure (kPa)
$P^0$	saturation pressure (kPa)
$R$	gas constant (cal/(g-mol K))
$T$	absolute temperature (K)
$W$	volume of the adsorbed phase given in eq 4 (cm <sup>3</sup> /(g of solid))
$W_0$	volume adsorbed at saturation (cm <sup>3</sup> /(g of solid))
$X$	amount adsorbed (g of adsorbate/(g of adsorbent))
$V$	amount adsorbed (cm <sup>3</sup> of adsorbate/(g of adsorbent))
$V_m$	amount adsorbed in the monolayer (cm <sup>3</sup> of adsorbate/(g of adsorbent))
$\beta$	affinity factor
$\epsilon$	adsorption potential (kcal/g-mol)

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## Adsorption of Synthesis Gas-Mixture Components on Activated Carbon

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Pure-gas isotherms have been determined for CH<sub>4</sub>, CO, CO<sub>2</sub>, and H<sub>2</sub> on type BPL activated carbon at 298.2 K by using a volumetric-type apparatus. Data for binary, ternary, and quaternary mixtures of these gases were obtained at the same temperature and a total pressure of 344.7 kPa. Two thermodynamic models which can predict gas-mixture equilibria from the pure-gas isotherms—the ideal adsorbed solution model (IASM) and the vacancy solution model (VSM)—have been evaluated with these data.

#### Introduction

There are several reasons for the success of activated carbon as an adsorbent. First, it can be prepared in a wide variety of structures having different pore sizes and pore volumes. Second, it exhibits high adsorption selectivity. For low boiling point compounds, the selectivity associated with activated carbon makes it an attractive alternative to other separational processes, such as fractional distillation which requires low temperatures and high pressures. Thus, activated carbon is used for such applications as the deodorization of air, the

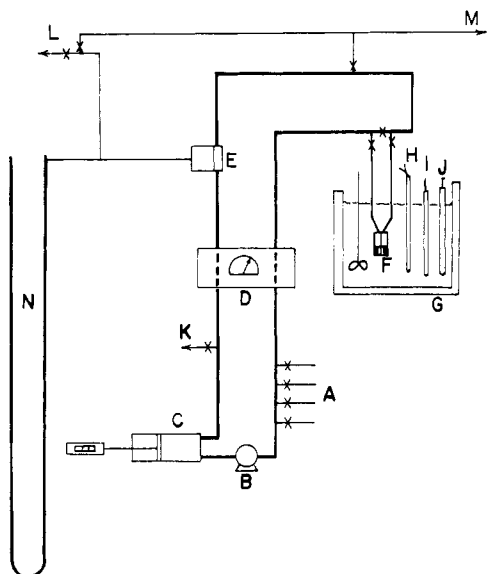


Figure 1. Schematic of the apparatus.

removal of ethylene from  $\text{CH}_4\text{-H}_2$  streams, the separation of isomers, and the purification of hydrogen. A brief but excellent review of the uses of activated carbon as an adsorbent has been published by Barneby (1).

The amount of literature that has been published on the adsorption of gases by solids is voluminous. Most of the research on activated carbon, however, has been confined to pure-gas adsorption (2-4). Only a few studies have been done on binary mixtures (5-7) while even less research has been done on ternary and higher-order mixtures (6, 8).

The primary objectives of this study were to enlarge the activated carbon data base for pure gases and gas mixtures of some of the major components found in synthesis gases and to evaluate the available models for predicting the mixture data from the pure-gas results.

## Experimental Section

**Apparatus.** The apparatus was of the volumetric type and used the difference technique for determining adsorbed-phase compositions. In this method the total quantity of each gas admitted to the system and the amount of each gas in the vapor remaining after adsorption equilibrium are determined by appropriate  $P$ - $V$ - $T$  measurements and analysis by a gas chromatograph. The adsorbed-phase parameters are then determined by the difference between the quantities of admitted and remaining gases. There are several important features of the equipment which enable a positive check on the attainment of equilibrium, accurate determinations of the equilibrium pressure without changes in the system volume, varying but precisely measured vapor-phase volumes, and the mixing of pure gases in situ.

A schematic diagram of the apparatus is shown in Figure 1. The recycle loop was constructed of  $1/4$ -in. o.d. copper tubing and brass fittings. A volumetric micrometer (C) permitted accurate measurement and adjustment of the internal volume of the recycle loop and simple regulation of the system pressure. It also served as a mixing chamber. A hermetically sealed, magnetically coupled centrifugal pump (B) provided circulation of the gases. Equilibrium was monitored by using a thermal conductivity detector (D) in which the thermal conductivities of streams entering and leaving the adsorption column were continuously compared. Attainment of adsorption equilibrium was indicated when the thermal conductivities of both streams were equal for the time equivalent to at least one recycle period. The adsorption column (F) was a brass cylinder with an

internal vertical baffle extending down below the level of the adsorbent sample. This assured intimate contact between the circulating adsorbates and the adsorbent. A three-valve manifold allowed isolation of the column and control over the direction of flow of the gases either through the adsorption column or through a bypass around the column.

A diaphragm type of differential-pressure cell (E) was used as a null pressure gauge. This permitted accurate measurements of the system pressure without any fluctuations in the system volume. Pressure measurements were made by admitting nitrogen gas to the reference side of the differential-pressure cell and by taking readings on a 60-in. mercury manometer (N). The adsorption column was submerged in a constant-level, constant-temperature bath of mineral oil (G). A proportional temperature controller coupled with an immersion heater regulated the bath temperature to within  $\pm 0.1$  K. Mechanical agitation of the bath liquid minimized temperature gradients. A gas chromatograph and an electronic digital integrator were used to analyze the equilibrium vapor phase.

The operating procedure for pure-gas isotherm determinations was to admit the gas into the recycle loop; measure its temperature, pressure, and volume; expand the gas into the adsorption column; and finally record the equilibrium temperature, pressure, and system volume. Changes in pressure could be made by admitting or removing known quantities of gas or by altering the system volume by using the volumetric micrometer.

In the gas-mixture experiments, multiple doses of pure gases were admitted, until a final equilibrium pressure of approximately 345 kPa was obtained. The order of admitting the pure gases was altered from one experiment to the next to be sure that no irreversible adsorption effects were being experienced.

The gases were circulated around the recycle loop until the thermal conductivity readings indicated that equilibrium was attained. The desired final pressure was maintained during the equilibration by adjusting the volumetric micrometer. The gas pump was shut off, and the static measurements were recorded. The column was isolated from the recycle loop, and vapor samples from the loop were analyzed in the gas chromatograph. Measurements for the residual gas in the loop were recorded, and further data points were taken by admitting more gases to the system and by repeating the equilibration procedure.

Between runs the adsorbent sample was regenerated in situ by heating to 573 K under a vacuum of approximately  $20 \mu\text{m}$  for a minimum of 12 h.

**Materials.** The adsorbent was type BPL activated carbon, a nonpolar, microporous, heterogeneous adsorbent, manufactured by Pittsburgh Activated Carbon, a division of the Calgon Corp. A surface area of  $1053 \text{ m}^2/\text{g}$  was determined by using the  $n$ -layers BET method (9). The dry weight of the carbon was determined by regenerating it at 530 K and less than  $10 \mu\text{m}$  for 45 h. The sample was then sealed and weighed immediately before it was transferred to the adsorption column. The CO had a minimum purity of 99.5%, the major impurities being  $\text{O}_2$  and  $\text{N}_2$ . Bone-dry  $\text{CO}_2$  and high-purity  $\text{H}_2$  (99.9999%) and  $\text{CH}_4$  (99.99%) were used.

## Data

**Pure-Gas Results.** Pure-gas isotherm data at 298.2 K were obtained for  $\text{CO}_2$ ,  $\text{CH}_4$ , CO, and  $\text{H}_2$  up to pressures of approximately 700 kPa (Figure 2 and 3). In all cases data were measured by using both adsorption and desorption steps. No hysteresis was observed.

None of the adsorbates is really polar although CO has a small permanent dipole (0.1 D) and  $\text{CO}_2$  has a sizable quadrupole moment ( $4.1 \times 10^{-26}$  esu  $\text{cm}^2$ ). In any case adsorption on the nonpolar carbon surface should not be strongly influ-

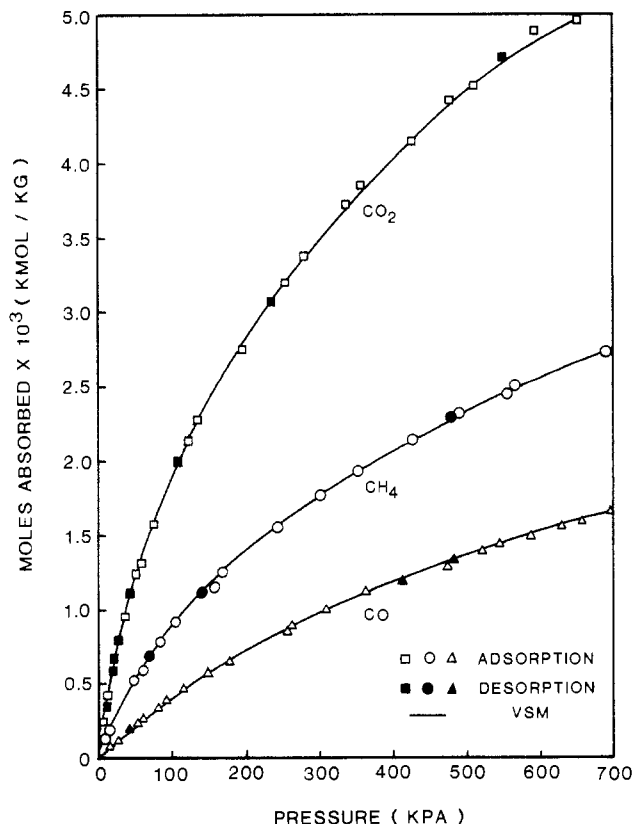


Figure 2. Carbon dioxide, methane, and carbon monoxide adsorption isotherms on BPL activated carbon at 298.2 K.

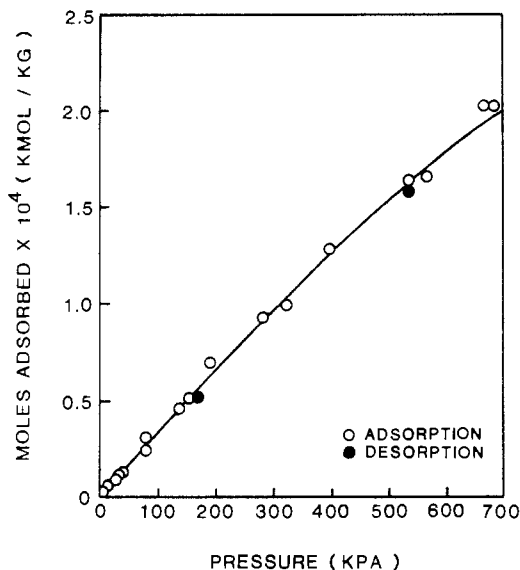


Figure 3. Hydrogen adsorption isotherm on BPL activated carbon at 298.2 K.

enced by any polar effects. The relative amounts adsorbed are in the same order as the critical temperatures or normal boiling points of the gases:  $H_2 < CO < CH_4 < CO_2$ . In other words, the more volatile substances are more weakly adsorbed. In these nonpolar, physical adsorption systems, there are no chemical or polar forces which would overcome the normal condensation behavior of these gases.

**Gas-Mixture Results.** Gas-mixture data for the  $CO-CO_2$ ,  $CO-CH_4$ , and  $CO_2-CH_4$  binary systems were obtained at 298.2 K and 344.7 kPa as listed in Table I. Adsorption phase diagrams and the total amount adsorbed for the  $CO-CO_2$  and  $CO-CH_4$  systems are shown in Figures 4 and 5. The selectivity is just as expected from the pure-gas adsorptivities with the

Table I. Binary Gas-Mixture Adsorption Data at  $P = 344.7$  kPa and  $T = 298.2$  K

$y_1$	$x_1$	$10^3 n_m^s$ , kmol/kg	$y_1$	$x_1$	$10^3 n_m^s$ , kmol/kg
Carbon Dioxide (1)-Carbon Monoxide (2)					
0.004	0.140	1.09	0.419	0.872	2.49
0.011	0.216	1.16	0.438	0.888	2.56
0.026	0.352	1.27	0.518	0.909	2.76
0.069	0.531	1.50	0.747	0.958	3.20
0.127	0.666	1.75	0.811	0.963	3.33
0.170	0.712	1.86	0.205 <sup>a</sup>	0.738	2.81
0.201	0.749	1.98	0.302 <sup>a</sup>	0.796	3.15
0.323	0.833	2.29	0.425 <sup>a</sup>	0.869	3.51
0.366	0.825	2.32			
Methane (1)-Carbon Monoxide (2)					
0.055	0.162	1.12	0.429	0.646	1.51
0.168	0.363	1.24	0.605	0.788	1.67
0.223	0.456	1.31	0.689	0.834	1.73
0.252	0.480	1.34	0.862	0.929	1.85
0.339	0.582	1.43			
Carbon Dioxide (1)-Methane (2)					
0.034	0.164	2.08	0.426	0.685	2.78
0.059	0.261	2.15	0.438	0.695	2.82
0.161	0.393	2.29	0.550	0.787	3.01
0.226	0.480	2.41	0.718	0.889	3.30
0.236	0.513	2.46	0.940	0.979	3.68

<sup>a</sup> Point at total pressure of 689.5 kPa.

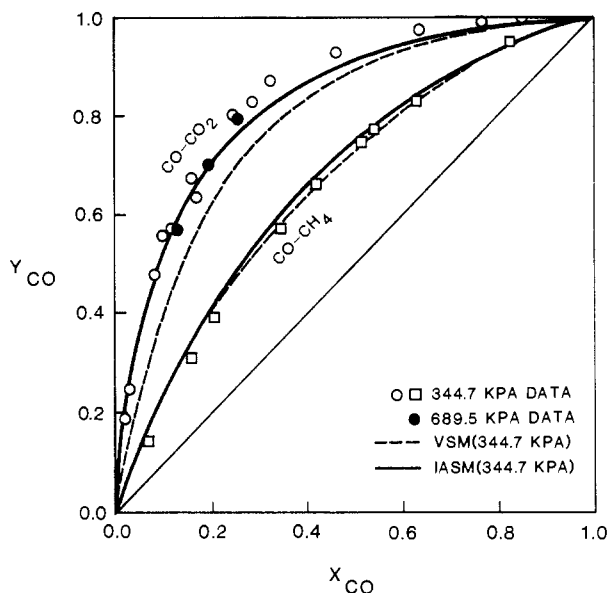


Figure 4. Adsorption phase diagram for  $CO-CO_2$  and  $CO-CH_4$  mixtures on BPL activated carbon at 298.2 K.

strongest separation appearing in the  $CO-CO_2$  system. Since the pure-gas adsorption of  $H_2$  is 1 order of magnitude smaller than the other gases, one would not expect it to compete well in binary systems with the other three gases. Data collected for the  $H_2-CO$  system confirmed this hypothesis. Within the experimental accuracy  $H_2$  acted only as a diluent; negligible  $H_2$  was adsorbed and the  $CO$  adsorption was equivalent to that obtained from the pure gas at a pressure equivalent to the partial pressure of  $CO$  in the  $H_2-CO$  mixture.

A few data points were collected for the  $CO-CO_2$  system at the same temperature but twice the pressure (689.5 kPa). No apparent change occurred in the phase compositions (Figure 4), but a significant increase in the total amount adsorbed was observed (Figure 5).

Eight data sets were obtained for ternary and quaternary mixtures at 298.2 K and 344.7 kPa total pressure. These data are listed in Table II. Hydrogen was treated as a vapor-phase diluent since no adsorption could be detected.

Table II. Gas-Mixture Adsorption Data and Prediction Results for Ternary and Quaternary Mixtures on BPL Activated Carbon at 298.2 K and 344.7 kPa Total Pressure

	IASM			VSM	
	exptl	calcd	% error	calcd	% error
$x_{CO_2}$	0.9204	0.9204	<i>a</i>	0.9204	<i>a</i>
$x_{CO}$	0.0274	0.0252	8.03	0.0274	<i>a</i>
$x_{CH_4}$	0.0522	0.0544	4.21	0.0522	<i>a</i>
$y_{CO_2}$	0.6957	0.6943	0.20	0.7428	6.77
$y_{CO}$	0.2008	0.1678	16.43	0.1285	36.00
$y_{CH_4}$	0.1035	0.1379	33.24	0.1268	22.51
$n_m^{s^4}$	0.003284	0.003271	0.40	0.003350	2.02
$x_{CO_2}$	0.9007	0.9007	<i>a</i>	0.9007	<i>a</i>
$x_{CO}$	0.0577	0.0552	4.33	0.0577	<i>a</i>
$x_{CH_4}$	0.0416	0.0441	6.01	0.0416	<i>a</i>
$y_{CO_2}$	0.5897	0.5836	1.03	0.6578	11.55
$y_{CO}$	0.3329	0.3205	3.72	0.2485	25.35
$y_{CH_4}$	0.0774	0.0959	23.90	0.0918	18.60
$n_m^{s^4}$	0.003031	0.002979	1.72	0.003188	5.17
$x_{CO_2}$	0.8068	0.8068	<i>a</i>	0.8068	<i>a</i>
$x_{CO}$	0.0258	0.0246	4.65	0.0258	<i>a</i>
$x_{CH_4}$	0.1674	0.1686	0.72	0.1674	<i>a</i>
$y_{CO_2}$	0.4942	0.5072	2.63	0.5453	10.34
$y_{CO}$	0.1646	0.1373	16.59	0.1058	35.72
$y_{CH_4}$	0.3412	0.3555	4.19	0.3469	1.67
$n_m^{s^4}$	0.002826	0.002919	3.27	0.003051	7.94
$x_{CO_2}$	0.7791	0.7791	<i>a</i>	0.7791	<i>a</i>
$x_{CO}$	0.0720	0.0784	8.88	0.0720	<i>a</i>
$x_{CH_4}$	0.1489	0.1425	4.30	0.1489	<i>a</i>
$y_{CO_2}$	0.3984	0.4008	0.60	0.4612	15.76
$y_{CO}$	0.3565	0.3552	0.37	0.2644	25.83
$y_{CH_4}$	0.2453	0.2440	0.53	0.2724	11.05
$n_m^{s^4}$	0.002581	0.002623	1.61	0.002855	10.61
$x_{CO_2}$	0.7256	0.7256	<i>a</i>	0.7256	<i>a</i>
$x_{CO}$	0.0586	0.0557	4.95	0.0586	<i>a</i>
$x_{CH_4}$	0.2158	0.2187	1.34	0.2158	<i>a</i>
$y_{CO_2}$	0.3666	0.3733	1.83	0.4094	11.67
$y_{CO}$	0.2716	0.2522	7.14	0.2089	23.09
$y_{CH_4}$	0.3618	0.3744	3.48	0.3798	4.98
$n_m^{s^4}$	0.002515	0.002616	4.01	0.002780	10.98
$x_{CO_2}$	0.7877	0.7877	<i>a</i>	0.7877	<i>a</i>
$x_{CO}$	0.0643	0.0692	7.62	0.0643	<i>a</i>
$x_{CH_4}$	0.1480	0.1430	3.33	0.1480	<i>a</i>
$x_{H_2}$	<i>a</i>	<i>a</i>	<i>a</i>	<i>a</i>	<i>a</i>
$y_{CO_2}$	0.3298	0.3405	3.24	0.3893	18.04
$y_{CO}$	0.2440	0.2632	7.87	0.1960	19.67
$y_{CH_4}$	0.1914	0.2061	7.68	0.2230	16.51
$y_{H_2}$	0.2348	0.2348	<i>a</i>	0.2348	<i>a</i>
$n_m^s$	0.002296	0.002676	16.53	0.002901	26.33
$x_{CO_2}$	0.8516	0.8516	<i>a</i>	0.8516	<i>a</i>
$x_{CO}$	0.0533	0.0531	0.38	0.0533	<i>a</i>
$x_{CH_4}$	0.0951	0.0953	0.21	0.0951	<i>a</i>
$x_{H_2}$	<i>a</i>	<i>a</i>	<i>a</i>	<i>a</i>	<i>a</i>
$y_{CO_2}$	0.5293	0.4466	15.62	0.5007	5.40
$y_{CO}$	0.1615	0.2468	52.82	0.1881	16.47
$y_{CH_4}$	0.1475	0.1673	13.42	0.1703	15.46
$y_{H_2}$	0.1617	0.1617	<i>a</i>	0.1617	<i>a</i>
$n_m^s$	0.003111	0.002876	7.54	0.003075	1.15
$x_{CO_2}$	0.8399	0.8399	<i>a</i>	0.8399	<i>a</i>
$x_{CO}$	0.0686	0.0688	0.29	0.0686	<i>a</i>
$x_{CH_4}$	0.0916	0.0913	0.33	0.0916	<i>a</i>
$x_{H_2}$	<i>a</i>	<i>a</i>	<i>a</i>	<i>a</i>	<i>a</i>
$y_{CO_2}$	0.4612	0.3940	14.57	0.4489	2.67
$y_{CO}$	0.2026	0.2851	40.72	0.2219	9.53
$y_{CH_4}$	0.1197	0.1430	19.47	0.1496	24.98
$y_{H_2}$	0.2165	0.2165	<i>a</i>	0.2165	<i>a</i>
$n_m^s$	0.002925	0.002778	5.03	0.003000	2.55

*a* Independent variable.

### Data Correlation and Prediction

A primary objective of research on the physical adsorption of gases is to allow multicomponent data to be predicted by using only pure-gas data. While pure-gas isotherms can be

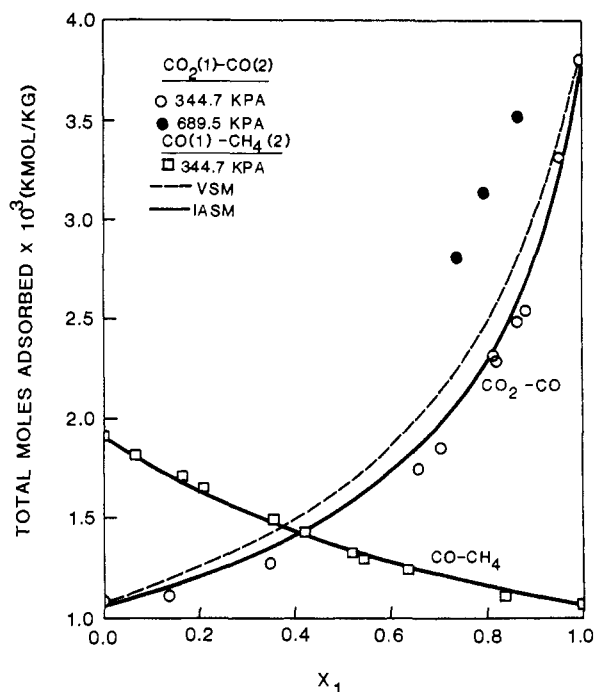


Figure 5. Total moles adsorbed from  $CO_2$ -CO and CO- $CH_4$  mixtures on BPL activated carbon at 298.2 K.

Table III. Vacancy Solution Model Regression Parameters

adsorbate	$10^3 n_1^{s,\infty}$ , kmol/kg	$b_1$ , kmol/(kg kPa)	$\Lambda_{1v}$	$\Lambda_{v1}$
$CO_2$	16.754	$4.665 \times 10^{-5}$	0.250	4.007
$CH_4$	10.857	$1.605 \times 10^{-5}$	0.269	3.713
CO	7.392	$5.32 \times 10^{-6}$	0.498	2.510
$H_2$	1.924	$3.377 \times 10^{-7}$	0.115	3.746

measured relatively easily, the measurement of gas-mixture adsorption equilibria has proved to be a tedious and expensive exercise. The present data sets should be useful to those who are attempting to develop new prediction techniques and provide an opportunity to test the currently available models. Previous work in our laboratories (10-14) has indicated that the ideal adsorbed solution model (15) and the vacancy solution model (14, 16) are the two most successful and versatile models for practical use. Both of these approaches are based on solution thermodynamic concepts.

The vacancy solution model (VSM) of Suwanayuen and Danner treats adsorption equilibria in terms of an equilibrium between a bulk vacancy solution and surface vacancy solution. The vacancies are hypothetical entities representing volume voids in the vapor phase or adsorbed phase which could be occupied by adsorbate molecules. Expressing the fugacity coefficient in each phase by the Wilson equation typically used in vapor-liquid equilibrium and equating the chemical potential of the adsorbate in the two phases leads to the VSM isotherm equation:

$$P = \left[ \frac{n_1^{s,\infty}}{b_1} \frac{\theta}{1-\theta} \right] \left[ \Lambda_{1v} \frac{1 - (1 - \Lambda_{v1})\theta}{\Lambda_{1v} + (1 - \Lambda_{1v})\theta} \right] \times \exp \left[ - \frac{\Lambda_{v1}(1 - \Lambda_{v1})\theta}{1 - (1 - \Lambda_{v1})\theta} - \frac{(1 - \Lambda_{1v})\theta}{\Lambda_{1v} + (1 - \Lambda_{1v})\theta} \right] \quad (1)$$

This equation has four parameters: the limiting adsorption amount,  $n_1^{s,\infty}$ , the Henry's law constant,  $b_1$ , and two Wilson equation parameters,  $\Lambda_{1v}$  and  $\Lambda_{v1}$ , which are interpreted as representing the interactions between the solid and adsorbate molecules. This equation correlates the pure-gas isotherms of

this work within the experimental accuracy as shown in Figures 2 and 3. The parameters are given in Table III. Unfortunately, for these volatile gases the relative pressure range covered is quite small even at 700 kPa and only the near-linear portions of the isotherms are available. Thus, the four parameters regressed on the basis of these poorly defined isotherms have questionable physical significance although they fit the isotherm data very well. In order to obtain more meaningful parameters, data are needed at very high pressures so that the value of  $n_i^{s,\infty}$  is defined by the data. Unfortunately, the present apparatus is not designed to operate in the high pressure range required.

For mixture predictions Suwanayuen and Danner (14) have shown that the VSM leads to an equation of the following form for each adsorbate:

$$\phi_i y_i P = \gamma_i^s x_i n_m^s \frac{n_i^{s,\infty} \Delta_{i,v}}{n_m^{s,\infty} b_i} \exp[\Delta_{v,i} - 1] \exp\left[\frac{\pi \bar{a}_i}{RT}\right] \quad (2)$$

Simultaneous solution of these equations together with the stipulations that  $\sum x_i = 1$  and  $\sum y_i = 1$  gives the mixture predictions desired. The VSM predictions for the CO-CO<sub>2</sub> and CO-CH<sub>4</sub> systems are shown in Figures 4 and 5. There is excellent agreement with the CO-CH<sub>4</sub> data and fair agreement with the CO-CO<sub>2</sub> data. Although not shown the predictions for the CO<sub>2</sub>-CH<sub>4</sub> system were also excellent.

For the ternary and quaternary systems the VSM gave errors in the phase compositions ranging from 1.7% to 36% and in the total moles adsorbed ranging from 1% to 26%. While these predictions leave room for improvement, they are not too bad considering that the isotherms from which the pure-component parameters were derived were not very well-defined.

The ideal adsorbed solution model (IASM) does not correlate pure-gas data. For gas mixtures it assumes ideal solution behavior between the vapor and adsorbed phases and, for each component, uses an expression analogous to Raoult's law in vapor-liquid equilibria.

$$P y_i = P_i^o(\pi) x_i \quad (3)$$

Equilibria are calculated by simultaneously solving this set of equations together with the requirements that  $\sum x_i = 1$  and  $\sum y_i = 1$ . To obtain the required spreading pressures, one integrates the pure-gas data from zero pressure to the pressure of interest. In the present case this was done by using the VSM equations.

The IASM did an excellent job of predicting all three binary systems. The CO-CO<sub>2</sub> and CO-CH<sub>4</sub> results are shown in Figures 4 and 5. For the ternary and quaternary systems the IASM gave errors in the phase compositions ranging from 0.2% to 53% and in the total moles adsorbed ranging from 0.4% to 16.5%.

## Conclusions

Extensive pure-component isotherm and binary adsorption equilibria data have been presented for CO, CO<sub>2</sub>, CH<sub>4</sub>, and H<sub>2</sub> on BPL activated carbon at 298.2 K. In addition, some ternary and quaternary data have been determined for these systems. The vacancy solution model correlates the pure-component data within experimental accuracy. Both the vacancy solution model and the ideal adsorbed solution model do a reasonable job of predicting the mixture equilibria data from the pure-com-

ponent isotherms. These systems are characterized by binary adsorption phase diagrams which are nearly symmetrical (i.e., ideal) and by pure-component isotherms which have not been determined over a sufficient range of pressure to give physical significance to the vacancy solution parameters. Thus, for these systems the ideal adsorbed solution model predictions are somewhat better than the vacancy solution model predictions. This is contrary to the results reported by Hyun and Danner (13) for azeotropic adsorption systems. In these nonideal systems the ideal adsorbed solution model could not predict azeotropes even qualitatively while the vacancy solution model did.

## Glossary

$\bar{a}_i$	partial molar area of adsorbate $i$ (m <sup>2</sup> /kmol)
$b_i$	Henry's law constant of adsorbate $i$ (kmol/(kg kPa))
$n_i^s$	number of moles of $i$ in surface phase (kmol/kg)
$n_i^{s,\infty}$	maximum number of moles of $i$ in surface phase (kmol/kg)
$n_m^s$	total number of moles of mixture in surface phase (kmol/kg)
$n_m^{s,\infty}$	maximum total number of moles of mixture in surface phase (kmol/kg)
$P$	pressure (kPa)
$P_i^o(\pi)$	pure-adsorbate adsorption pressure at spreading pressure, $\pi$ (kPa)
$R$	universal gas constant (m <sup>3</sup> kPa/(kmol K))
$T$	temperature (K)
$x_i$	mole fraction of component $i$ in adsorbed phase
$y_i$	mole fraction of component $i$ in vapor phase

## Greek Letters

$\gamma_i^s$	activity coefficient of $i$ in adsorbed-phase vacancy solution
$\theta$	fractional coverage, $n_i^s/n_i^{s,\infty}$
$\Delta_{i,v}, \Delta_{v,i}$	Wilson's interaction parameters
$\pi$	spreading pressure (N/m)
$\phi_i$	fugacity coefficient of $i$ in bulk gas mixture

**Registry No.** C, 7440-44-0; CH<sub>4</sub>, 74-82-8; CO, 630-08-0; CO<sub>2</sub>, 124-38-9; H<sub>2</sub>, 1333-74-0.

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