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T. Victor Lee[a]; Richard Madey[a]; Jan-Chan Huang[b]
[a] DEPARTMENT OF PHYSICS, KENT STATE UNIVERSITY, KENT, OHIO [b] DEPARTMENT OF PLASTICS ENGINEERING, UNIVERSITY OF LOWELL, LOWELL, MASSACHUSETTS

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Adsorption Equilibria for Ethane and Propane Gas Mixtures on Activated Carbon

T. VICTOR LEE and RICHARD MADEY

DEPARTMENT OF PHYSICS
KENT STATE UNIVERSITY
KENT, OHIO 44242

JAN-CHAN HUANG

DEPARTMENT OF PLASTICS ENGINEERING
UNIVERSITY OF LOWELL
LOWELL, MASSACHUSETTS 01854

Abstract

We measured the transmission curves of binary gas mixtures of ethane and propane (and also of each gas alone) flowing through an adsorber bed at 25°C packed with (Columbia 4LXC 12/28) activated carbon. The adsorption isotherms for pure ethane and propane are described well by a modified Langmuir isotherm known as the Chakravarti-Dhar isotherm. Ethane in the binary mixtures exhibits smaller adsorption capacities than that of pure ethane; however, the presence of ethane does not significantly reduce the adsorption capacity of propane in the mixtures. The ideal adsorbed solution theory (IAST) of Myers and Prausnitz and the semiempirical method of Cook and Basmadjian (C-B) are applied to predict adsorption equilibrium properties of the binary mixtures from the known pure gas isotherms. The pure ethane isotherm data at pressures higher than 10 mmHg are obtained from a correlation formula. Good agreement between the predicted results from the IAST and C-B methods and the experimental results indicates (1) that the correlation formula can be used for estimating isotherms of related hydrocarbons with reliance and (2) that the two methods are useful in predicting the adsorption equilibrium properties for the ethane-propane mixtures on carbon.

INTRODUCTION

Gas adsorption on solid adsorbents such as activated carbons, molecular sieves, and polymers is an attractive process for the separation

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of gases. In practice, the gas adsorption process involves the treatment of multicomponent mixtures. The design of an adsorption separator is based on a knowledge of equilibrium properties.

In this paper the ideal adsorbed solution theory (IAST) of Myers and Prausnitz (*1*) and the semiempirical method of Cook and Basmadjian (*2*) will be applied to predict the adsorption equilibrium properties for binary gas mixtures of ethane and propane from the known pure gas isotherms. The predictions will be compared with dynamic measurements of the transmission of ethane and propane and their binary mixtures through an (Columbia 4LXC 12/28) activated carbon adsorber bed at 25°C. A correlation formula (*3*) is used to extrapolate the adsorption isotherm of ethane for pressures above 10 mmHg where experimental data are lacking.

EXPERIMENTAL METHOD

A gas chromatographic method was used to measure the transmission of an adsorbate gas through an adsorber bed. Transmission is the ratio of the concentration at the outlet of the bed to that at the inlet. Since the flow rate system used in the present study was described elsewhere (*4*), only a brief description is given here. The flow system was constructed of stainless-steel tubing and valves with Teflon seats and gaskets. Calibrated hydrocarbon–helium mixtures (with a nominal concentration of 1%) were prepared by the Matheson Gas Co. (East Rutherford, New Jersey). The calibrated gases were mixed with pure helium in different proportions. Gas flow rates were measured by flowmeters with an accuracy of ±0.5%. The concentration of the hydrocarbon at the downstream side of the column was measured at regular time intervals by a Varian 3700 gas chromatograph.

The cylindrical stainless-steel adsorber bed (10.02 cm long and 0.459 cm i.d.) was packed with 0.588 g of "Columbia" type 4LXC 12/28 activated carbon. This carbon has intrinsic density 1.560 g/cm^3 (*5*), surface area 1,130 m^2/g (6), and pore volume 0.51 cm^3/g (*6*). Before measuring the mass of the carbon, the adsorber bed was desorbed at 200°C with helium flowing through the adsorber bed at a rate of 200 cm^3/min for 36 h. Also, between each run the bed was desorbed for 12 h under the same temperature and helium flow conditions as above. The bed was immersed in a constant-temperature water bath which maintained the temperature within ±0.02°C at 25°C. Repeated experiments verified the reproducibility of the results.

The equilibrium solid-phase concentration q_0 can be calculated from a mass-balance equation (7) when the time-dependent transmission C/C_0 is known.

$$q_0(1 - \varepsilon)L + C_0\varepsilon L = uC_0\varepsilon \int_0^\infty (1 - C/C_0)dt \tag{1}$$

Here the adsorbent void fraction ε includes the voids within the carbon granules, L is the length of the column, u is the superficial flow velocity, and C is the concentration of the trace component measured at the column exit.

In our experiment, helium is used as the carrier gas. Since the adsorption capacity of helium is negligible at room temperature, the partial pressure of helium is not included for further data manipulations.

TWO PREDICTIVE METHODS FOR BINARY ADSORPTION EQUILIBRIA

Two methods for predicting binary adsorption equilibria that will be used in this paper for evaluating the experimental binary data are: (a) the ideal adsorbed solution theory (IAST) of Myers and Prausnitz (*1*) and (b) the semiempirical method of Cook and Basmadjian (*2*). Summaries of the two methods are given below.

Ideal adsorbed solution theory is based on the two-dimensional thermodynamics of the (solid) adsorbed phase. The essential assumption is that the mixtures adsorbed on solid adsorbents form an ideal adsorbed solution (viz., activity coefficient $\gamma_i = 1$) at a constant spreading pressure π; that is, for each component i, the gas partial pressure equals the product of the solid-phase mole fraction X_i and the equilibrium pressure $P_i^\circ(\pi)$ which is a function of the spreading pressure π:

$$P_T Y_i = P_i = X_i P_i^\circ(\pi) \tag{2}$$

The Gibbs adsorption equation (1) relates the spreading pressure π to the measurable pure gas isotherm:

$$\frac{\pi_i A}{RT} = \int_0^P \frac{n_i^\circ}{P_i} dP_i \tag{3}$$

where A is the specific surface area of the adsorbent, R is the universal gas constant, and T is the absolute temperature. The pure gas isotherm relates the solid-phase concentration n_i° to the gas-phase concentration which is expressed in terms of the partial pressure P_i. IAST also describes an ideal adsorbed phase as a mixture which has no surface change or enthalpy change upon mixing at constant temperature and constant spreading pressure. The criterion of zero surface change leads to the following expression for a binary mixture:

$$a = a_1^\circ(\pi)X_1 + a_2^\circ(\pi)X_2 \tag{4}$$

where a, a_1°, and a_2° denote the molar surface area of the mixture, component 1, and component 2, respectively. Since the molar surface area a_i° is inversely proportional to the amount n_i° of adsorbate on the adsorbent, Eq. (4) can be rewritten as:

$$\frac{1}{n_T} = \frac{X_1}{n_1^\circ(\pi)} + \frac{X_2}{n_2^\circ(\pi)} \tag{5}$$

When the single-component isotherm [viz., $n_i^\circ(\pi)$ from Eq. 3] and the solid-phase mole fraction (viz., X_i obtained from Eq. 2) are known, the total amount n_T of the binary gas mixture adsorbed on the solid adsorbent can be determined by Eq. (5). Then the amount of individual gas component adsorbed in the solid phase can be obtained from the following mass-balance equation:

$$n_i = X_i n_T \tag{6}$$

Cook and Basmadjian (*2*) proposed a semiempirical method for the prediction of binary adsorption equilibria from pure gas isotherms. The C-B method consists of evaluating the separation factor at two extremes of infinite dilution and interpolating the values at intermediate compositions through an empirical procedure. The formulas that relate the two limiting separation factors to the pure gas isotherms are:

$$\log \alpha_1 = \frac{1}{n_1^\circ} \int_0^{n_1^\circ} \log\left(\frac{P_1^0}{P_2^\circ}\right)_n dn \tag{7}$$

and

$$\log \alpha_2 = \frac{1}{\overset{\circ}{n}_2} \int_0^{\overset{\circ}{n}_2} \log \left(\frac{P_1^0}{P_2^\circ} \right)_n dn \tag{8}$$

Here the separation factor α is defined by $\alpha \equiv (Y_1/X_1)/(Y_2/X_2)$, and α_1 is the value of α as $X_1 \rightarrow 1$ (and $Y_1 \rightarrow 1$), and α_2 is the value of α as $X_2 \rightarrow 1$ (and $Y_2 \rightarrow 1$). When the heavy component (viz., component 2) dominates (i.e., $X_2 \rightarrow 1$), Y_2/X_2 approaches unity and $\alpha_2 = Y_1/X_1$; thus, when $X_2 \rightarrow 1$,

$$P_T\alpha_2 = P_T(Y_1/X_1) = P_1/X_1 \tag{9}$$

Similarly, when $X_1 \rightarrow 1$,

$$P_T/\alpha_1 = P_T(Y_2/X_2) = P_2/X_2 \tag{10}$$

The total amount of gas adsorbed (n_T) and the ratio of the partial pressure to the solid-phase mole fraction (i.e., P_i/X_i) are connected semiempirically by a linear relationship. For each value of n_T between $\overset{\circ}{n}_1$ and $\overset{\circ}{n}_2$, values of P_1/X_1 and P_2/X_2 can be interpolated with the use of the boundary conditions stated in Eqs. (9) and (10). One of the unknown mole fractions, for example X_2, can be determined by solving the following semiempirical formula of Cook and Basmadjian (*2*):

$$X_2 = \frac{\left(\dfrac{P_1}{X_1} \right)_{n_T} - P_T}{\left(\dfrac{P_1}{X_1} \right)_{n_T} - \left(\dfrac{P_2}{X_2} \right)_{n_T}} \tag{11}$$

The other adsorption equilibrium properties (viz., X_1, Y_1, Y_2, P_1, P_2, n_1, and n_2) can be obtained from mass balance equations.

RESULTS

The experimental pure gas isotherms for ethane and propane on (Columbia 4LXC 12/28) activated carbon at 25°C are plotted in Fig. 1. For each of the two gases, the equilibrium solid-phase concentration n is

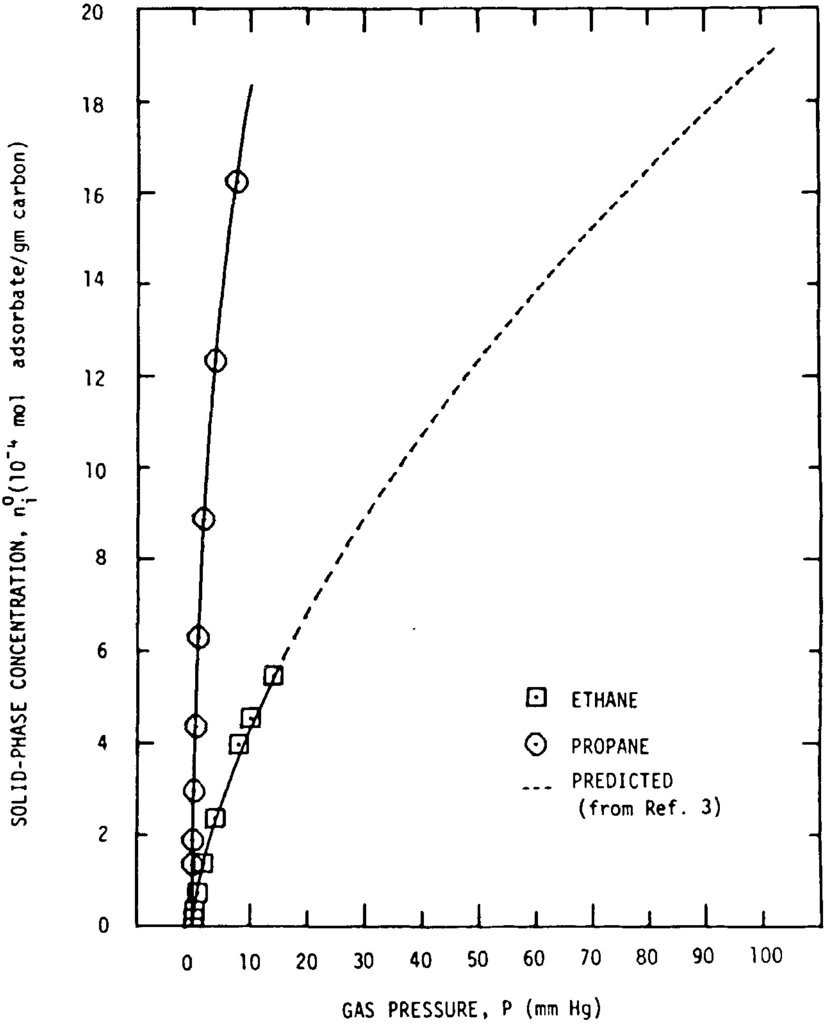

FIG. 1. Adsorption isotherms for the pure gases ethane and propane on Columbia 4LXC 12/28 activated carbon at 25°C.

not linearly proportional to the equilibrium gas-phase concentration $C(= P/RT)$; however, the adsorption isotherms can be represented by a modified Langmuir isotherm known as the Chakravarti-Dhar (*8*) isotherm; that is,

$$\frac{n}{n_m} = \frac{(K_m C)^{\nu}}{1 + (K_m C)^{\nu}} \tag{12}$$

where n_m is the solid-phase concentration for a monolayer coverage and K_m and ν are constants. Equation (12) simplifies to the Langmuir isotherms when $\nu = 1$ and to the Freundlich equation (*9*) when K_m is small. The ethane isotherm in Fig. 1 consists of the experimental points represented by the symbols and the extrapolated region represented by the dashed line. The extrapolated region is obtained from our previous correlation formula (*3*). The correlation is based on the Polanyi-Dubinin adsorption theory (*10*) and the use of the Peng-Robinson (*11*) equation of state to determine the thermodynamic properties. In order to apply the two predictive methods for calculating adsorption equilibrium properties of binary gas mixtures, it is necessary to know the adsorption isotherm for each of the pure gases in the two-component mixture at the same solid-phase concentration; thus, it is necessary to extend the adsorption isotherm for ethane to a pressure range above that of the present experimental data. A computer code CURFIT (*12*), which makes a least-squares fit to a nonlinear function, was used to determine the parameters of the Chakravarti-Dhar type isotherm for ethane and propane. The results are listed in Table 1. The small values of reduced chi-square, χ_{ν}^2, indicate that the deviations between the experimental and calculated adsorption isotherm data (viz., the solid-phase concentration n_i°) are small. The fact that the observed probability $P_{\chi}(\chi^2, \nu)$ to the assumed Chakravarti-Dhar equation is nearly unity means Eq. (12) is a good fit to the experimental isotherm of ethane and propane. The probability $P_{\chi}(\chi^2, \nu)$ is the probability of exceeding χ^2 at the number of degrees of freedom ν (*12*).

The solid-phase concentrations for each component and for the sum of the two components in the ethane–propane binary mixtures are plotted in Fig. 2 versus the gas-phase mole fraction of ethane. Although the five experimental data points in Fig. 2 were obtained at slightly different gas pressures, the small variation from 7.64 to 7.75 mmHg does not significantly affect our analysis. The gas mixtures in our experiments were prepared by mixing the carrier gas (helium) with the calibrated gas mixtures while flowing through the adsorber bed. It was difficult to maintain the partial pressure of the gas mixtures at a fixed value.

TABLE 1

Parameters and Statistics of Chakravarti-Dhar Type Isotherms for Ethane and Propane on Columbia 4LXC 12/28 Activated Carbon at 25°C

		Parameter			Statistics		
Gas	Highest gas pressure (mmHg)	K_m (10^5 cm^3/mol)	n_m (10^{-1} mol/cm^3)	v	Reduced chi-square χ_ν^2	Probability $P_\chi(\chi^2, \nu)$	Number of data points
Ethane	100	0.168	0.189	0.703	0.155×10^{-5}	≃ 1.00	19
Propane	10	1.91	0.120	0.516	0.168×10^{-6}	≃ 1.00	8

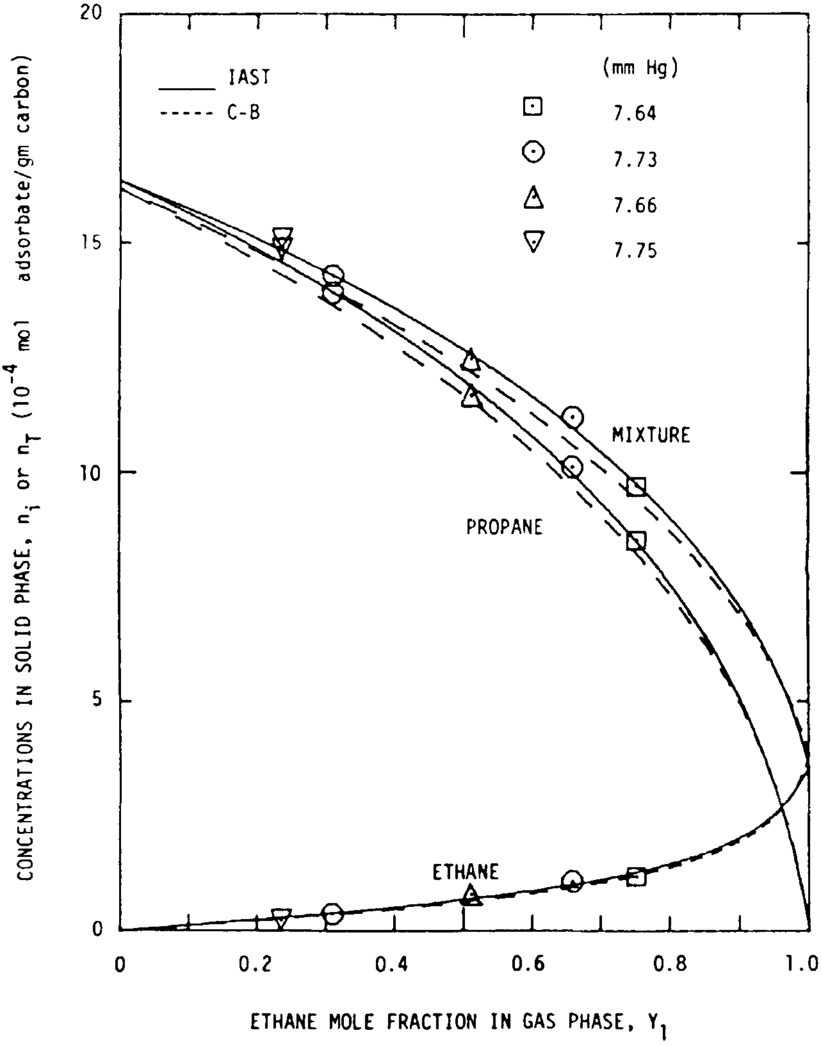

FIG. 2. Adsorbed amount of binary mixtures of ethane and propane on Columbia 4LXC 12/28 activated carbon at 25°C versus the mole fraction of ethane in the gas phase at a total pressure of 7.7 mmHg.

The predicted solid-phase concentrations from the C-B method are within a 5% deviation of those obtained from experiments. The values calculated from the IAST method give a better result with deviations less than 2%. Numerical values for the experimental and predicted solid-phase concentrations are listed in Table 2.

The logarithm of the solid-phase mole fraction versus the gas-phase mole fraction for ethane is plotted in Fig. 3. The solid-phase mole fractions calculated by both the IAST and the C-B methods agree with those from experiment. The largest absolute deviation is 0.009 for the C-B method and is 0.008 for the IAST. The relative deviations in solid-phase mole fractions are less than 1.2% for propane but range up to 20% for ethane. The relative deviations are larger for ethane than for propane because the solid-phase mole fractions are much smaller for ethane. The values of the separation factor calculated from Eqs. (7) and (8) are $\alpha_1 = 27.7$ and $\alpha_2 = 17.5$ at a pressure of 7.7 mmHg and 25°C for ethane-propane mixtures.

DISCUSSION

In gas-solid adsorption of a binary gaseous mixture, the two gas components will affect each other as they are adsorbed on the solid phase. The dashed lines in Figs. 4 and 5 show the amount of ethane and propane adsorbed from mixtures of the two gases on activated carbon as a function of the gas pressure. For comparison, the individual pure gas isotherms are represented by the solid lines in the two figures. It is seen in Fig. 4 that over the whole pressure range the adsorption of ethane is depressed by the presence of propane whereas, as shown in Fig. 5, the adsorption of propane is not significantly affected by the presence of ethane. Previously, Madey, Forsythe, and Huang (*13*) studied the dynamic transport of gas mixtures of ethane and propane through an adsorber bed of activated carbon and found that a large portion of the adsorbed ethane is displaced by the later-eluting propane. The interference and displacement phenomena result from interactions between the two gases in the solid phase. The result from the dynamic study is consistent with the results obtained here.

We applied the IAST to predict the equilibrium properties of binary mixtures of ethane and propane on activated carbon and obtained good agreement with the experimental results; however, a question of how far the adsorption behavior of these binary mixtures deviate from the assumption of the ideal adsorbed solution theory (viz., $\gamma_i = 1$) arises.

TABLE 2

Experimental and Calculated Solid-Phase Concentration for Mixtures of Ethane and Propane at a Pressure of about 7.7 mmHg on Columbia 4LXC 12/28 Activated Carbon at 25°C

Gas-phase pressure (mmHg), P_T	Gas-phase concentration (10^{-7} mol/cm^3)		Total solid-phase concentration for mixture, n_T (10^{-4} mol/g carbon)[a]		
	Ethane, C_1	Propane, C_2	Experimental	IAST (Δ)	C-B method (Δ)
7.64	3.09	1.02	9.71	9.72 (+0.13)	9.39 (−3.3)
7.73	2.74	1.42	11.2	11.0 (−1.8)	10.6 (−5.1)
7.66	2.11	2.01	12.5	12.5 (+0.50)	12.1 (−2.4)
7.73	1.29	2.87	14.3	14.3 (+0.22)	14.0 (−2.1)
7.75	0.99	3.18	15.1	14.9 (−1.4)	14.6 (−3.5)

[a] $\Delta \equiv 100[(n_t)_{calc} - (n_T)_{expt}]/(n_T)_{expt}$.

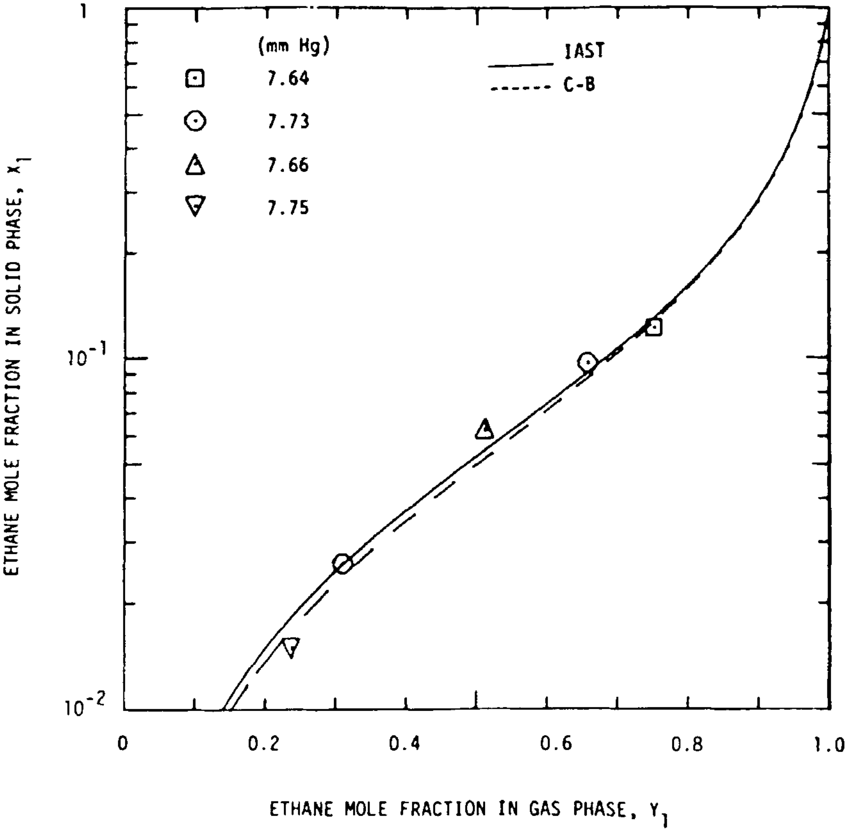

FIG. 3. Equilibrium phase diagram for adsorption of binary mixtures of ethane and propane on Columbia 4LXC 12/28 activated carbon at 25°C at a total pressure of 7.7 mmHg.

When we take into account the activity coefficient, Eq. (2) can be rewritten as

$$P_T Y_i = \gamma_i X_i P_i^\circ(\pi) \tag{13}$$

where X_i represents the experimental solid-phase mole fraction for the *i*th component. Combination of Eqs. (2) and (13) gives

$$1 \cdot (X_i)_{\text{IAST}} = \gamma_i (X_i)_{\text{EXPT}} \tag{14}$$

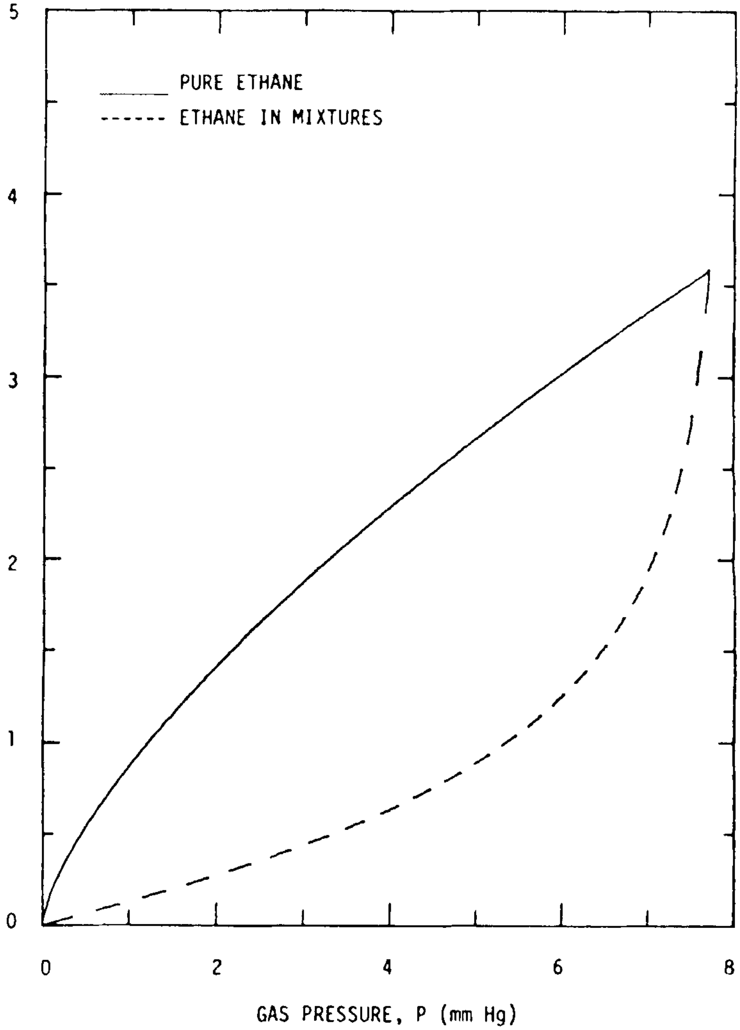

FIG. 4. Solid-phase concentration of ethane on Columbia 4LXC 12/28 activated carbon at 25°C versus the gas pressure for pure ethane and for ethane in binary mixtures of ethane and propane at a total pressure of 7.7 mmHg.

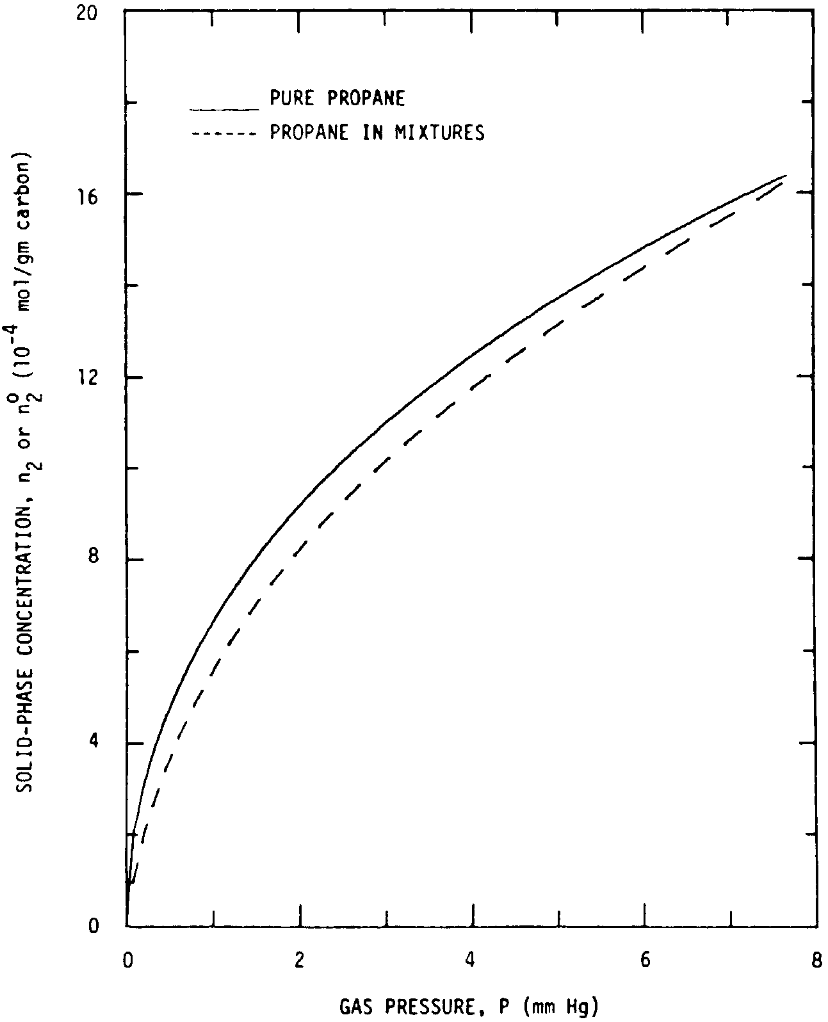

FIG. 5. Solid-phase concentration of propane on Columbia 4LXC 12/28 activated carbon at 25°C versus the gas pressure for pure propane and for propane in binary mixtures of ethane and propane at a total pressure of 7.7 mmHg.

or

$$\gamma_i = \frac{(X_i)_{\mathrm{IAST}}}{(X_i)_{\mathrm{EXPT}}} \tag{15}$$

The activity coefficients calculated by Eq. (13) or Eq. (15) are summarized in Table 3. For propane the values of the activity coefficient are close to unity (viz., $0.989 < \gamma_2 < 1.01$). The activity coefficients for ethane deviate from unity (viz., $0.87 < \gamma_1 < 1.22$). From mass-balance equations we know that the absolute deviations between the IAST and the experimental solid-phase mole fractions [viz., $|(X_i)_{\mathrm{IAST}} - (X_i)_{\mathrm{EXPT}}|$] are identical for the two components in a binary mixture. Since the heavy-component propane dominates in the solid phase, the activity coefficients, which are the ratios of $(X_i)_{\mathrm{IAST}}$ to $(X_i)_{\mathrm{EXPT}}$, are close to unity. On the other hand, the activity coefficients for the depressed-component ethane show large deviations from unity. With activity coefficients being close to unity, propane is considered as an ideal component in the binary mixtures, while ethane behaves less ideally as far as the activity coefficient is concerned. From the results shown in Table 2 and Fig. 2, appreciable deviations of the activity coefficient from unity for the depressed-component ethane do not affect the ability of the IAST to predict the total solid-phase concentration of the mixtures with accuracy. As seen in Table 3, the prediction of the adsorbed-phase mole fraction of the depressed-component ethane deviates from experiment by an amount comparable with the deviation of the activity coefficient from unity.

Previously (*3*), we proposed a formula for estimating the isotherms of pure hydrocarbon gases. In this paper we proceed to use the estimated isotherms to predict the adsorption equilibrium properties of binary mixtures. The estimated isotherm of ethane, together with the experimental isotherms for each individual gas, were used to calculate the adsorption equilibrium properties for ethane–propane mixtures. Results calculated from IAST (shown in Fig. 2 and Table 2) are in excellent agreement with the experimental ones, and those calculated from the C-B method also agree but with somewhat larger errors. With the success of this procedure, it is demonstrated that our correlation formula can be used to determine the isotherms for related hydrocarbons with reliance.

CONCLUSION

We measured the transmission of pure gases (viz., ethane and propane) and binary mixtures of these gases in a helium carrier gas flowing

TABLE 3

Activity Coefficients for Mixtures of Propane (Component 1) and Ethane (Component 2) at a Pressure of 7.7 mmHg Columbia 4LXC 12/28 Activated Carbon at 25°C

Gas-phase mole fractions, Experimental		Adsorbed-phase mole fractions, Experimental		Adsorbed-phase mole fractions, IAST Method		Equilibrium gas concentration (mmHg)		Activity coefficient	
Ethane, Y_1	Propane, Y_2	Ethane, X_1	Propane, X_2	Ethane, $\bar{X}_1$	Propane, $\bar{X}_2$	Ethane, $P_1^\circ(\pi)$	Propane, $P_2^\circ(\pi)$	Ethane, γ_1	Propane, γ_2
0.752	0.248	0.122	0.878	0.130	0.870	44.2	2.18	1.06	0.989
0.659	0.341	0.097	0.903	0.091	0.909	55.8	2.90	0.941	1.01
0.512	0.488	0.063	0.937	0.055	0.945	71.6	3.95	0.870	1.00
0.310	0.690	0.026	0.974	0.026	0.974	92.4	5.47	0.997	1.00
0.237	0.763	0.015	0.985	0.018	0.982	99.9	6.03	1.22	0.994

through an (Columbia 4LXC 12/28) activated carbon adsorber bed at 25°C. The adsorption isotherms for both single-component gases are nonlinear and can be represented by a modified Langmuir isotherm known as the Chakravarti-Dhar isotherm.

We applied the ideal adsorbed solution theory (IAST) and the Cook and Basmadjian (C-B) method to calculate the equilibrium properties of gas–solid adsorption for binary mixtures of ethane and propane at a pressure of about 7.7 mmHg on activated carbon at 25°C from the corresponding single-component isotherms. Both the IAST and C-B methods provide close predictions to the experimental results; thus, both methods are considered useful in predicting the equilibrium properties of ethane–propane mixtures.

The work of this paper suggests that the isotherms of light hydrocarbon gases calculated from a previous correlation formula (*3*) can be used with reliance. It becomes convenient that the availability of a generalized correlation on the isotherms of related substance (e.g., hydrocarbons) will enable one to apply existing predictive models to obtain the equilibrium properties for the adsorption of binary and/or multicomponent gas mixtures.

Acknowledgment

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SYMBOLS

A	specific surface area for an adsorbent (cm^2/g)
a	molar surface area for binary mixtures (cm^2/mol of adsorbate mixture)
a_i°	molar surface area for the pure component i (cm^2/mol of the ith component)
C	gas-phase concentration at the exit of the column (mol/cm^3)
C_i	gas-phase concentration for the pure gas i (mol/cm^3)
C_0	gas-phase concentration at the inlet of the column (mol/cm^3)
K_m	constant in Eq. (12) (cm^3/mol)
L	length of the adsorber column (cm)

n	solid-phase concentration used in Eq. (12) (mol/cm^3)
n_i	solid-phase concentration for the component gas i (mol/cm^3)
n_i°	solid-phase concentration for the pure gas i (mol/cm^3)
n_m	equilibrium solid-phase concentration for a monolayer coverage in Eq. (12) (mol/cm^3)
n_T	solid-phase concentration for binary mixtures (mol/cm^3)
P_i	partial pressure for the ith component gas in mixture (mmHg)
P_T	total pressure for binary gas mixtures (mmHg)
P_i°	gas pressure for the pure gas i (mmHg)
$P_{(\chi^2,\nu)}$	probability of exceeding χ^2 at the number of degrees of freedom ν
q_0	equilibrium solid-phase concentration in an adsorber column (mol/cm^3)
R	universal gas constant (1.987 cal/°K·mol)
T	absolute temperature (°K)
t	time (s)
u	superficial flow velocity (cm/s)
X_i	mole fraction of the ith component in the solid phase (dimensionless)
Y_i	mole fraction of the ith component in the gas phase (dimensionless)

Subscripts

EXPT	experimental method
IAST	ideal adsorbed solution theory
i	ith component (i = 1, 2 for binary mixture)
m	monolayer coverage
0	initial or inlet concentration of a column
T	total quantity for mixture
1	(light) component 1
2	(heavy) component 2

Superscript

$^\circ$	Pure gas or substance

Greek Letters

α	separation factor $\equiv (Y_1/X_1)/(Y_2/X_2)$ (dimensionless)
α_1	α as $X_1 \rightarrow 1$ (dimensionless)
α_2	α as $X_2 \rightarrow 1$ (dimensionless)
ε	void fraction of adsorber bed (dimensionless)
γ_i	activity coefficient of component i in the solid phase (dimensionless)
π_i	spreading pressure of gas component i (dyn/cm)
ν	constant in Eq. (12) (dimensionless)
χ_ν^2	reduced chi-square (dimensionless)

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