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An Improved Langmuir Approach for Prediction of Binary Gas Mixture Adsorption Equilibria*

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ABSTRACT

The mixed Langmuir adsorption isotherm has been modified for improved prediction of binary gas mixture adsorption equilibria. The modified method has been applied to predict adsorption equilibria for 52 binary gaseous mixtures on adsorbents like activated carbon, silica gel, and carbon and zeolite molecular sieves for which experimental data are reported in the literature. Predictions by this method are generally better for the binary gas mixtures in systems involving activated carbons, silica gel, and carbon molecular sieve adsorbents compared to that of the mixed Langmuir approach. However, in systems having zeolite molecular sieve adsorbents, predictions are less satisfactory. The higher deviations of predicted data from experimental values in these systems are attributed to the lack of single component adsorption isotherm data at low coverages.

INTRODUCTION

Design and optimization of an adsorptive gas separation process invariably require data on multicomponent gas adsorption. However, literature on such data is sparse because measurement of these data is laborious and involves tedious experimentation. Hence, prediction schemes using only pure component adsorption data for gas-mixture equilibria are generally used. Numerous efforts (1–3) made in the past several years for developing theoretical models for the prediction of multicomponent adsorption equilibria from the pure gas adsorption equilibria of the components have

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resulted in models such as the ideal adsorbed solution theory (4, 5), the vacancy solution model (6), the adsorption potential model (7–11), and the simplified statistical thermodynamic model (12–17). Despite being fundamentally strong, these models are rarely used in practice for design and optimization purposes on a commercial scale. This is essentially because all these models involve three or four parameters, thus necessitating enormous computer time during the design and optimization of a process. As a result, the Langmuir isotherm extended for multicomponent gas mixtures, which has only two parameters, remains the most widely practiced model (3) in commercial applications although it is not always satisfactory. The errors in the prediction of multicomponent adsorption by the mixed Langmuir largely comes from the Langmuir constant b which is obtained from single component isotherms. In the present paper we report a method wherein the use of individual b values is avoided, thereby improving the predictions of adsorption equilibria.

THEORY

On extending the Langmuir adsorption isotherm to binary gas mixtures, we obtain

$$V_1 = \frac{(V_m)_1 b_1 p_1}{1 + \sum b_1 p_1 + b_2 p_2} \tag{1}$$

where p_i are the partial pressures and b_i are the Langmuir constants for Component i, V_i are the amounts adsorbed from the mixture, and $(V_m)_i$ are the monolayer volumes for Component i.

Similarly for Component 2:

$$V_2 = \frac{(V_m)_2 b_2 p_2}{1 + \sum b_1 p_1 + b_2 p_2} \tag{2}$$

The total volume adsorbed for a binary gas mixture is given by

$$V = V_1 + V_2 = \frac{(V_m)_1 b_1 p_1 + (V_m)_2 b_2 p_2}{1 + b_1 p_1 + b_2 p_2} \tag{3}$$

Dividing Eq. (1) by Eq. (3), we obtain the adsorbed phase composition with respect to Component 1:

$$x_1^a = \frac{(V_m)_1 b_1 p_1}{(V_m)_1 b_1 p_1 + (V_m)_2 b_2 p_2} \tag{4}$$

or

$$x_1^a = \frac{1}{1 + \dfrac{(V_m)_2}{(V_m)_1} \dfrac{b_2}{b_1} \dfrac{p_2}{p_1}} \tag{5}$$

For a given gas phase composition, x_1^a can be calculated from Eq. (5) if the values of the Langmuir constants for the two gases are known.

The experimental values of b and V_m for single gases are determined from the intercept and slope of the Langmuir plot. The intercept of the Langmuir plot is often small and hence may not be determined accurately. Consequently, the value of b as obtained from the Langmuir plot is normally subject to error. However, for the prediction of adsorption equilibrium data, as seen from Eq. (5), the ratio b_2/b_1 is necessary and not the absolute values of b_1 and b_2.

At present, the ratio b_2/b_1 is determined accurately from the following considerations. At a given concentration range for the two gases (or at a given adsorbed volume if the values of V_m for the two gases are close), we have

$$\theta = \frac{b_1 p_1}{1 + b_1 p_1} = \frac{b_2 p_2}{1 + b_2 p_2} \tag{6}$$

or

$$\frac{1}{b_1 p_1} = \frac{1}{b_2 p_2} \tag{7}$$

or

$$\frac{b_2}{b_1} = \frac{p_1}{p_2} \tag{8}$$

Thus, the ratio b_2/b_1 can be obtained from the ratio of the equilibrium pressures of the individual adsorption isotherms of the two gases at the same adsorption loading.

It has to be pointed out, however, that the ratio b_2/b_1 has to be determined at low pressures for the following reasons. The constants b_1 and b_2 are related to the energies of adsorption of the respective gases. The adsorption from mixtures is largely determined by the relative magnitude of the adsorption energies. The adsorbate with a higher adsorption energy is selectively adsorbed. In many adsorbents, like zeolite molecular sieves, the surface coverage is quite high, even at low pressure. It is the adsorption energy in this region that determines the mixture adsorption, and therefore it is necessary that the ratio b_2/b_1 should be determined in a low pressure region. Thus the ratio of b_2/b_1 ($= p_1/p_2$ as shown in Eq. 8) was obtained at several coverages in the $\theta = 0.2$ to 0.5 range from individual isotherms. For many systems the ratio b_2/b_1 was observed to vary with adsorption loading. In such systems, the value of b_2/b_1 used for prediction was obtained by extrapolating to zero coverage.

RESULTS AND DISCUSSION

The improved Langmuir method has been used to predict the adsorption phase diagrams for 52 binary systems for which experimental data were taken from the literature. These systems have been broadly divided into four groups based on the type of adsorbent: silica gel, activated carbon, carbon molecular sieves (CMS), and zeolite molecular sieves (ZMS). The source and the characteristics of adsorbents are given in Table 1. The

TABLE 1
Details of Various Adsorbents Included in the Present Study and Method of Adsorption Measurement

Adsorbent	Source	Surface area (m^2/g)	Particle size	Method of adsorption measurement
Silica gel	Davision Chemical Co.,	751		Static volumetric
Activated carbon fiber KF-1500	Toyobo Co.			Static volumetric
Activated carbon AC-40	Compania Espain la de Carbones Actives S.A.	700	1.66 mm diameter, 4.26 mm height	Volumetric
Activated carbon Nuxit-Al			1.5 mm diameter, 3–4 mm long	Static volumetric
Activated carbon EY-51-C	Pittsburg Coke Co.	805		Static volumetric
Carbon molecular sieve, CMS-5A	Takeda Chemical Co.	650	28–42 mesh	Volumetric
Carbon molecular sieve, CMS-3A	Takeda Chemical Co.			Volumetric
ZMS, 5A	Linde, UCC		1.5 mm diameter pellets with 20% binder	Volumetric
ZMS, 10X	Linde, UCC		1.5 mm diameter pellets with 20% binder	Volumetric
ZMS, 13X (Hyun)	Linde, UCC	525	1.5 mm diameter pellets with 20% binder	Static volumetric
ZMS, 13X (Bhat)	Linde, UCC		1.5 mm diameter pellets with 20% binder	Volumetric
ZMS, H-mordenite	Norton Co., type Z-900H		1.5 mm diameter pellets	Static volumetric

various adsorbates include permanent gases such as N_2, O_2, Ar, CO, CO_2, and light hydrocarbons. The Langmuir constants V_m and b for single component have been calculated for the above systems by linear regression of the experimental data into the Langmuir isotherm. The Langmuir constants thus obtained, along with the standard deviation (σ) and the relative mean square deviation (s), as determined by Eqs. (9) and (10), are given in Tables 2–5.

$$\sigma = \sqrt{\sum (V_{\mathrm{cal}} - V_{\mathrm{exp}})^2/n} \tag{9}$$

$$s = \sqrt{\frac{\sum \left(\frac{V_{\mathrm{cal}} - V_{\mathrm{exp}}}{V_{\mathrm{exp}}}\right)^2}{n}} \tag{10}$$

where V_{exp} and V_{cal} are experimental and calculated equilibrium adsorption capacities, and n is the number of data.

Adsorption equilibrium data for binary mixtures have been predicted for 10 silica gel systems, 18 activated carbon systems, 7 carbon molecular sieve systems, and 19 zeolite molecular sieve systems. For comparison purpose, the adsorption equilibrium data for the above binary mixtures have also been predicted using the mixed Langmuir equation. The average relative deviation of predicted adsorbed phase composition from the experimental data in each system is presented in Tables 6–9. The ratios of V_{m2}/V_{m1} and the values b_2/b_1 used in the improved Langmuir approach are also given in these tables.

Predicted adsorption data by the present improved Langmuir method (Table 6) for binary systems having silica gel adsorbent show relative

TABLE 2
Langmuir Constants for Various Adsorbates on Silica Gel

Adsorbent	Adsorbate	T (K)	V_m (mol·kg^{-1})	b (kPa^{-1})	Standard deviation	rms deviation	Ref.
Silica gel	Acetylene	298.15	2.1236	0.036264	0.072	0.137	27
Silica gel	Ethylene	278.15	2.0663	0.026251	0.042	0.102	25
Silica gel	Ethylene	298.15	1.8718	0.009927	0.012	0.039	27
Silica gel	Ethylene	313.15	1.2758	0.011350	0.016	0.086	25
Silica gel	Propane	273.15	3.4712	0.023177	0.086	0.098	25
Silica gel	Propane	298.15	2.2664	0.017545	0.052	0.139	24
Silica gel	Propane	313.15	1.9452	0.011578	0.026	0.096	25
Silica gel	Propylene	273.15	3.7254	0.049315	0.079	0.048	25
Silica gel	Propylene	298.15	2.7099	0.034929	0.073	0.130	27
Silica gel	Propylene	313.15	2.2826	0.030427	0.053	0.065	25

TABLE 3
Langmuir Constants for Various Gases on Activated Carbons

Adsorbent	Adsorbate	T (K)	V_m (mol·kg^{-1})	b (kPa^{-1})	Standard deviation	rms deviation	Ref.
AC (Nuxit-Al)	Methane	293.15	2.4523	0.004935	0.014	0.031	23
AC (Nuxit-Al)	Ethane	293.15	3.7353	0.049958	0.122	0.149	23
AC (Nuxit-Al)	Ethane	333.15	2.7919	0.021144	0.045	0.056	23
AC (Nuxit-Al)	Ethylene	293.15	3.6178	0.035180	0.096	0.122	23
AC (Nuxit-Al)	Ethylene	333.15	2.7161	0.016166	0.042	0.054	23
AC (Nuxit-Al)	Propane	293.15	4.3492	0.242471	0.279	0.301	23
AC (Nuxit-Al)	Propane	333.15	3.5528	0.083610	0.155	0.208	23
AC (Nuxit-Al)	Propylene	293.15	4.7383	0.150355	0.277	0.237	23
AC (Nuxit-Al)	*n*-Butane	293.15	4.0018	0.510208	0.297	0.313	23
AC (Nuxit-Al)	CO_2	293.15	4.2065	0.010349	0.039	0.063	23
AC (EY-51-C)	Acetylene	298.15	2.6508	0.027672	0.076	0.113	27
AC (EY-51-C)	Ethylene	298.15	2.6448	0.034350	0.058	0.073	27
AC (EY-51-C)	Propane	298.15	2.7352	0.094895	0.125	0.125	27
AC (EY-51-C)	Propylene	298.15	2.9226	0.088933	0.103	0.084	27
AC40	Methane	293.15	1.0501	0.011035	0.046	0.099	18
AC40	Ethane	293.15	2.3992	0.066617	0.125	0.257	18
AC40	Ethylene	293.15	2.1957	0.050086	0.097	0.193	18
AC40	Propylene	293.15	3.2975	0.196000	0.231	0.357	18
Activated carbon fiber	Methane	298.15	6.5824	0.001786	0.069	0.035	20
Activated carbon fiber	CO_2	298.15	11.8678	0.002611	0.209	0.075	20

TABLE 4
Langmuir Constants for Various Gases on Carbon Molecular Sieves

Adsorbent	Adsorbate	T (K)	V_m (mol·kg^{-1})	b (kPa^{-1})	Standard deviation	rms deviation	Ref.
CMS-3A	Nitrogen	298.15	1.3954	0.002308	0.034	0.112	19
CMS-3A	Oxygen	298.15	1.3926	0.002502	0.016	0.069	19
CMS-5A	Ethylene	274.85	2.6566	0.271152	0.147	0.150	16
CMS-5A	Ethylene	303.15	1.9186	0.238523	0.066	0.109	16
CMS-5A	Ethylene	323.15	1.8793	0.123150	0.051	0.097	16
CMS-5A	Propylene	274.85	2.7020	1.118613	0.235	0.123	16
CMS-5A	Propylene	303.15	2.2328	1.115928	0.096	0.069	16
CMS-5A	Propylene	323.15	2.1136	0.618069	0.115	0.106	16
CMS-5A	Ethane	278.65	2.6040	0.246700	0.146	0.147	28
CMS-5A	Ethane	303.15	2.2515	0.151802	0.079	0.101	28
CMS-5A	Ethane	323.15	2.0552	0.094546	0.064	0.104	28
CMS-5A	Propane	278.15	3.0396	0.962687	0.353	0.328	28
CMS-5A	Propane	303.15	2.8700	0.802535	0.204	0.265	28
CMS-5A	Propane	323.15	2.7304	0.499062	0.160	0.248	28

TABLE 5
Langmuir Constants for Various Gases on Zeolite Molecular Sieves

Adsorbent	Adsorbate	T (K)	V_m (mol·kg^{-1})	b (kPa^{-1})	Standard deviation	rms deviation	Ref.
5A	Oxygen	144.26	5.4679	0.118733	0.107	0.103	21
5A	Nitrogen	144.26	4.6244	0.744077	0.335	0.216	21
5A	CO	144.26	4.9876	1.690197	0.634	0.183	21
10X	Oxygen	144.26	6.7536	0.028065	0.105	0.149	21
10X	Nitrogen	144.26	4.9025	0.184405	0.274	0.252	21
10X	CO	144.26	5.8138	0.285301	0.512	0.275	21
13X	Ethylene	298.15	2.8639	0.277231	0.119	0.185	15
13X	Ethylene	323.15	2.5928	0.118057	0.090	0.127	15
13X	Ethylene	373.15	2.0777	0.030238	0.042	0.087	15
13X	Ethane	298.15	2.8983	0.033410	0.057	0.075	15
13X	Ethane	323.15	3.1594	0.011653	0.034	0.050	15
13X	Ethane	373.15	3.8485	0.002399	0.006	0.028	15
13X	Isobutane	298.15	1.8732	0.646962	0.297	0.308	15
13X	Isobutane	323.15	1.6449	0.598227	0.164	0.190	15
13X	Isobutane	373.15	1.3819	0.263107	0.032	0.086	15
13X	CO_2	298.15	4.0972	0.169633	0.213	0.240	15
13X	CO_2	323.15	3.6395	0.079474	0.141	0.175	15
13X	Argon	89.15	7.1352	6.357961	0.859	0.247	22
13X	Oxygen	89.15	7.4509	6.977140	1.148	0.327	22
13X	Nitrogen	89.15	6.6334	7.582572	1.164	0.261	22
13X	CO	89.15	6.9741	16.736464	0.735	0.171	22
H-mordenite	CO_2	303.15	2.4600	0.081228	0.143	0.251	26
H-mordenite	H_2S	303.15	2.3368	0.640899	0.162	0.147	26
H-mordenite	Propane	303.15	1.2157	0.237615	0.131	0.354	26

TABLE 6
Deviation of the Predicted Adsorbed Phase Composition by Simplified Langmuir and Mixed Langmuir Methods for Silica Gel Systems

			Relative average deviation (%)		Maximum deviation (%)				
Gas mixture	T (K)	No. data	Improved Langmuir	Mixed Langmuir	Improved Langmuir	Mixed Langmuir	V_{m1}/V_{m2}	b_2/b_1	Ref.
Propylene–propane	298.15	12	29	28	78	79	0.836	0.470	24
Propylene–ethylene	273.15	8	26	34	63	71	1.680	0.419	25
Propylene–ethylene	298.15	6	23	29	44	54	0.836	0.230	25
Propylene–ethylene	313.15	5	14	35	23	54	0.852	0.200	25
Propane–ethylene	273.15	12	9	11	26	30	1.803	1.070	25
Propane–ethylene	298.15	9	10	14	28	38	1.448	0.619	25
Propane–ethylene	313.15	6	2	2	4	5	1.789	0.967	25
Acetylene–ethylene	298.15	6	11	22	44	71	1.135	0.340	25

TABLE 7
Deviation of the Predicted Adsorbed Phase Composition by Simplified Langmuir and Mixed Langmuir Methods for Activated Carbon Systems

			Relative average deviation (%)		Maximum deviation (%)				
Gas mixture	T (K)	No. data	Improved Langmuir	Mixed Langmuir	Improved Langmuir	Mixed Langmuir	V_{m1}/V_{m2}	b_2/b_1	Ref.
Adsorbent: AC-40:									
Methane–ethylene	293.15	5	19	43	45	95	0.481	6.750	18
Methane–ethane	293.15	6	28	60	63	95	0.440	10.500	18
Ethylene–ethane	293.15	6	9	9	18	18	0.915	1.340	18
Ethylene–propylene	293.15	6	17	78	31	111	0.666	11.100	18
Ethane–propylene	293.15	6	12	71	21	108	0.728	7.800	18
Adsorbent: AC(EY-51-C):									
Ethylene–acetylene	298.15	10	12	11	29	32	0.998	0.830	27
Propylene–propane	298.15	20	8	8	36	37	1.069	1.083	27
Adsorbent: Carbon fiber KF-1500:									
Methane–CO_2	298.15	6	25	28	45	50	0.555	1.520	20
Adsorbent: Nuxit-Al:									
Propylene–propane	293.15	7	5	23	13	35	1.089	1.120	23
Propylene–ethylene	293.15	5	3	27	9	46	1.309	0.116	23
Propane–ethane	293.15	10	4	9	15	22	1.164	0.136	23
Propane–ethane	333.15	5	2	5	3	15	1.273	0.194	23
Ethylene–methane	293.15	7	4	5	10	12	1.476	0.132	23
Ethylene–CO_2	293.15	6	3	3	7	5	0.860	0.282	23
Ethane–methane	293.15	7	4	5	13	14	1.523	0.097	23
Ethane–ethylene	293.15	10	3	1	5	3	1.032	0.738	23
Ethane–ethylene	333.15	7	3	3	5	4	1.028	0.777	23
n-Butane–propane	293.15	7	7	28	24	44	0.920	0.247	23

TABLE 8
Deviation of the Predicted Adsorbed Phase Composition by Simplified Langmuir and Mixed Langmuir Methods for Carbon Molecular Sieve Systems

			Relative average deviation (%)		Maximum deviation (%)				
Gas mixture	T (K)	No. data	Improved Langmuir	Mixed Langmuir	Improved Langmuir	Mixed Langmuir	V_{m1}/V_{m2}	b_2/b_1	Ref.
Adsorbent: CMS-3A:									
Nitrogen–oxygen	298.15	22	26	28	85	86	1.001	1.024	19
Adsorbent: CMS-5A:									
Ethylene–propylene	274.85	11	43	84	88	186	0.656	8.300	16
Ethylene–propylene	303.15	8	33	50	80	75	0.573	8.500	16
Ethylene–propylene	323.15	10	20	56	39	140	0.593	9.700	16
Ethane–propane	278.15	6	31	53	65	84	0.857	9.690	17
Ethane–propane	303.15	11	23	29	63	47	0.791	7.550	17
Ethane–propane	323.15	7	25	34	68	52	0.756	9.080	17

TABLE 9
Deviation of the Predicted Adsorbed Phase Composition by Simplified Langmuir and Mixed Langmuir Methods for Zeolite Molecular Sieve Systems

			Relative average deviation (%)		Maximum deviation (%)				
Gas mixture	T (K)	No. data	Improved Langmuir	Mixed Langmuir	Improved Langmuir	Mixed Langmuir	V_{m1}/V_{m2}	b_2/b_1	Ref.
Adsorbent: 5A:									
Oxygen–nitrogen	144.26	11	60	19	83	38	1.182	25.000	21
Oxygen–CO	144.26	12	52	48	72	112	1.096	59.800	21
Nitrogen–CO	144.26	12	114	253	268	641	0.928	5.250	21
Adsorbent: 13X:									
Nitrogen–argon	89.15	15	61	53	97	96	0.930	1.340	22
Argon–oxygen	89.15	20	35	55	61	77	0.958	1.580	22
Nitrogen–CO	89.15	9	117	115	202	198	0.951	2.100	22
Adsorbent: 10X:									
Oxygen–nitrogen	144.26	11	49	26	75	51	1.378	13.158	21
Nitrogen–CO	144.26	10	69	152	173	402	0.843	2.950	21
Oxygen–CO	144.26	11	43	26	63	64	1.162	20.000	21
Adsorbent: 13X:									
Isobutane–ethylene	298.15	10	34	20	66	45	0.654	0.710	15
Isobutane–ethylene	323.15	8	34	20	62	26	0.634	0.124	15
Isobutane–ethylene	373.15	6	22	26	51	65	0.665	0.128	15
Isobutane–ethane	298.15	10	28	12	121	56	0.646	0.019	15
Isobutane–ethane	323.15	7	6	23	16	86	0.521	0.043	15
Ethylene–CO_2	298.15	6	29	39	42	78	0.699	0.940	15
Ethylene–CO_2	323.15	5	19	14	31	26	0.658	0.615	15
Adsorbent: H-mordenite:									
CO_2–propane	303.15	9	40	55	84	91	2.023	1.600	26
H_2S–CO_2	303.15	9	3	13	8	36	0.950	0.068	26
H_2S–propane	303.15	8	27	42	46	71	1.922	0.170	26

average deviations from 2% for propane–ethylene at 313.15 K to 29% for propylene–ethylene at 298.15 K. It is also observed that mixtures having propylene as one of the components show higher deviations. However, as seen from Table 6, deviations from the mixed Langmuir isotherm are generally on the high side compared to those with the improved approach. For example, the relative average deviation for the binary mixtures propylene–ethylene at 313.15 K and acetylene–ethylene at 298.15 K by mixed Langmuir are double compared to those predicted by the improved Langmuir equation.

In almost all the systems involving activated carbon adsorbent, the adsorption equilibrium data predicted by improved Langmuir method are in excellent agreement with the experimental data. Predictions by the present method compare very well with the experimental data of Szepesy and

Illes (23) on activated carbon Nuxit-Al with almost negligible deviations. Further, data predicted by the present method compare well with the data of Costa et al. (18) except in mixtures having methane as one of the components. Of the 18 systems for which the adsorption data are predicted, in as many as 12 binary mixtures the relative average deviation is less than 10%, as seen from Table 7. Only two systems showed relative average deviations of more than 20% (methane–ethane at 293.15 K and methane–CO_2 at 298.15 K showed 28 and 25% deviations, respectively). On the other hand, in the majority of systems the errors in predictions by the mixed Langmuir equation are on the high side. In the case of mixed Langmuir equations, the relative average deviations are as high as 78%.

In systems having carbon molecular sieve 5A, prediction by the improved Langmuir method is fairly good (relative average deviations 23 to 31%) in the binary systems of nitrogen–oxygen at 298.15 K and ethane–propane at 278.15, 303.15, and 323.15 K as observed in Table 8. However, deviations of data predicted by the present approach from that of experimental data for ethylene–propylene systems at 274.85, 303.15, and 323.15 K reported by Nakahara et al. (16) are high, with the highest deviation at 274.85 K. High scatter in experimental mixture data, particularly at low ethylene loadings, may be one of the reasons for high deviations in the predicted data. However, deviations in predictions by the mixed Langmuir method are higher in these systems than those predicted by the improved method.

The improved Langmuir approach has been applied to predict the adsorption equilibria for 19 binary mixtures with zeolites as adsorbents. The data are given in Table 9. In general, deviations in predictions in these systems are high compared to systems having silica gel or activated carbon as adsorbents. Only in three cases (isobutane–ethane on 13X at 323.15 K, ethylene–CO_2 at 323.15 K, and H_2S–CO_2 on H-mordenite) are the predictions comparable to those of experimental data, and the relative average deviations in these systems are less than 20%. Deviations are more than 50% in 7 systems. It is observed that some gases are very strongly adsorbed in zeolites even at very low pressures, making it difficult to measure adsorption isotherms accurately at low coverages. Whenever experimental data on a single component are not available at low coverages, the predictions of binary mixture adsorption are not very accurate. In the majority of systems used in the present study, a single component adsorption isotherm for one of the gases was reported at adsorption loadings above 0.4. For example, carbon monoxide, which possesses both dipole (0.12 esu $\times$ 10^{10}) and quadrupole moments (0.33 Å^3), interacts strongly with the zeolite surface, and the pure component experimental data for carbon monoxide on zeolites are not available below $\theta = 0.4$. Consequently the ratio b_2/b_1 was obtained at higher coverages (above 0.4) and then extrapolated to zero loading graphically. As a result, all the systems containing carbon monoxide as one of the components gave 50 to 117% deviations. Hence, it can be concluded that the higher deviations in the case of zeolite molecular sieves are mostly due to lack of experimental single component isotherm data in the very low pressure region (i.e., at low coverages).

The improved Langmuir method described in the present paper is generally better than the mixed Langmuir approach for binary gases on activated carbons, silica gel, and carbon molecular sieve adsorbents. The predictions are not very satisfactory for systems involving zeolite molecular sieve adsorbents. This is largely due to the lack of availability of single component adsorption isotherm data at low coverages.

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