Comparing Various Multicomponent Adsorption Equilibrium Models

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In this article effects of energy distribution function and load multicomponent isotherm on predictions of multicomponent adsorption equilibria are studied. The effect of the single-component isotherm equation on the ideal adsorbed solution theory (IAST) predictions was also investigated. Five predictable multicomponent adsorption equilibrium models were evaluated using binary and ternary experimental data in the literature. Two of the models apply the IAST at a patch of sites having uniform energy to yield a local adsorption isotherm, and then the local isotherm is integrated over the full energy distribution assumed to take either uniform or binomial functional form. The third model uses a local extended Langmuir equation and the integration is done over a uniform energy distribution. The last two methods assume the global IAST with the single-component isotherm described by either the Unilan or Toth equation. The prediction results depend on the choice of energy distribution function and the single-component isotherm equation. In general, the heterogeneous IAST seems to be a better approach than a global IAST.

The prediction of multicomponent adsorption equilibria using single-component isotherm information is still one of the most challenging problems in adsorption studies. The prediction results of the multicomponent adsorption equilibrium significantly affect the multicomponent sorption kinetics model predictions. An error in the multicomponent equilibrium prediction can cause a larger error in the dynamics prediction (Hu and Do, 1992), and there have been many attempts to tackle this problem. The simplest method is to use an extended Langmuir equation to describe the multicomponent equilibria. The advantage of this method is that the expression for equilibria is analytical and explicit, hence making the dynamic numerical computation much faster. The disadvantage of this approach is that the extended Langmuir equation is inadequate representing the real adsorption system, because the Langmuir equation cannot fully describe the single-component adsorption data and/or the maximum adsorption capacity differs from species to species, causing thermodynamic inconsistency. To overcome this consistency, Myers and Prausnitz (1965) proposed an ideal adsorbed solution theory (IAST). The goodness of the IAST in predicting multicomponent adsorption equilibrium depends on the correct choice of the single-component isotherm equations best describing experimental data (Myers, 1987; Richter et al., 1989; Hu and Do, 1992). The deviations from ideal behavior are often attributed to the nonideality of the adsorbed phase. There are two approaches in dealing with this nonideality. One attributes the nonideality to a result from nonunity activity coefficients of the solution, and it is called real adsorbed solution theory (RAST) (Costa et al., 1981; Talu and Zwiebel, 1986). The RAST has extra parameters to be extracted from binary system, hence it requires binary experimental data in order to predict higher-order multicomponent equilibria. Another approach considers the surface energetic heterogeneity, inaugurated by Roginski (1948) for single-component systems. Since the original article by Roginski was published in Russia, this approach only became widely known after the work of Ross and Olivier published in 1961. Hoory and Prausnitz (1967) further extended the energy distribution to multicomponent mixtures. In dealing with multicomponent adsorption equilibria, this methodology assumes that IAST is applicable at a local site and the overall adsorption isotherm can be integrated over all available sites, through the use of an energy distribution. This is referred as heterogeneous ideal adsorbed solution theory (HIAST). Since different sorbates have different energy distribution toward the solid surface, Myers (1987) and Valenzuela et al. (1988) used the matching of the cumulative energy of all species. With this approach, the prediction on multicomponent adsorption equilibria would require no extra fitting parameter. Moon and Tien (1988) used the same theory based on regular or complementary site-matching technique, with binary data needed to find some parameters. Another approach in multicomponent equilibrium prediction is the heterogeneous extended Langmuir (HEL) method proposed by Kapoor et al. (1990). It has an advantage that the solution is explicit with reasonably good results if the maximum adsorption capacities are the same for all species (Hu and Do, 1993) or are close to each other (Kapoor et al., 1990). However, it has a similar drawback as that in the extended Langmuir equation, that is, the thermo-

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dynamical consistency when the maximum adsorption capacity differs significantly from species to species. In this article the effects of surface heterogeneity, choice of single-component isotherm equation, local multicomponent isotherm, and energy distribution function on the multicomponent equilibria predictions will be explored. This is achieved by comparing binary and ternary equilibria predictions using information of single-component isotherm data from five different models along with experimental results. Two of the models use the IAST with a Toth or Unilan as the single-component isotherm equation, respectively. The third model is the HEL and the last two methods utilize the HIAST and a binomial (Valenzuela et al., 1988) or uniform energy distribution (this study). The binomial function is a discrete one in nature. The effect of single-component isotherm parameter extraction procedure on the multicomponent adsorption equilibrium predictions will also be discussed.

Theory

If $E(k)$ is the adsorbate-adsorbent interaction energy at a patch of sites having uniform energy and $F_k[k, E(k)]$ is the energy distribution in the adsorbed phase for species k, respectively, the observed adsorbed phase concentration C_μ is

$$C_\mu(k) = \int_0^\infty C_\mu[k, E(k)] F_k[k, E(k)] dE(k) \qquad (1)$$

where $C_\mu[k, E(k)]$ is the adsorbed phase concentration at a given energy site and its relationship with the gas-phase concentration is usually called a local adsorption isotherm. In this study the local isotherm is assumed to satisfy the ideal adsorbed solution condition and is calculated from the IAST. The single-component adsorption equilibrium equation used in the local IAST computations is assumed to take a Langmuir form:

$$C_\mu[k, E(k)] = C_{\mu s}(k) \frac{b_0(k) \exp[E(k)/R_g T] p(k)}{1 + b_0(k) \exp[E(k)/R_g T] p(k)} \qquad (2)$$

where p is the gas-phase concentration, $C_{\mu s}$ is the maximum adsorption capacity, b_0 is the affinity constant at zero energy level, R_g is the gas constant, and T is temperature in Kelvin.

A uniform energy distribution is assumed in this article, which takes the following form:

$$F_k[k, E(k)] = \frac{1}{E_{\max}(k) - E_{\min}(k)} \quad \text{for} \quad E_{\min} < E < E_{\max} \qquad (3)$$

and the function is zero elsewhere.

The ordering of the adsorption sites from low to high and the cumulative energy distribution are generally assumed the same for each adsorbate species (Valenzuela et al., 1988). When these assumptions were applied to a uniform distribution, the energy relationship for different adsorbates is given by Kapoor et al. (1990):

$$\frac{E(i) - E_{\min}(i)}{E_{\max}(i) - E_{\min}(i)} = \frac{E(j) - E_{\min}(j)}{E_{\max}(j) - E_{\min}(j)} \qquad (4)$$

The local IAST equations are available in Valenzuela et al. (1988) and readers are referred to their article for more details about IAST.

Results and Discussion

In the last section we have used an IAST model to describe the local multicomponent equilibrium, and with a uniform energy distribution we derived the overall multicomponent adsorption equilibrium. This theory will be applied to analyze experimental data available in the literature. The results from the same HIAST utilized a discrete binomial energy distribution (Valenzuela et al., 1988) that will also be reported and compared to show the effect of the choice of energy distribution function in the prediction of multicomponent adsorption equilibria. Three other models to be compared are: the heterogeneous extended Langmuir (HEL) model (Kapoor et al., 1990), and two global IAST models with a Unilan or Toth equation used to describe the single-component isotherm. The results from HIAST-uniform and IAST-Unilan will reveal the effect of applying IAST locally or globally on the prediction of multicomponent equilibria. The two global IAST models using a single-component isotherm equation of either Unilan or Toth will give information about the choice of single-component isotherm in the calculation of IAST. It is noted that in the five models the HIAST-uniform, the HEL and the IAST-Unilan have the same single-component adsorption equilibrium isotherm, which is the Unilan equation. The differences between them are in the treatment of heterogeneity in predicting equilibria of multicomponent systems.

First the data reported by Talu and Zwiebel (1986) were reanalyzed. Figure 1 shows the single-component adsorption equilibrium isotherm of CO_2, H_2S, and C_3H_8 on the hydrogen mordenite. The semi-log plot was used to clearly show the adsorption equilibria at low pressure. The experimental data are represented as symbols, the isotherm fitting using a local Langmuir equation and a uniform energy distribution (Unilan equation) as solid lines. In parameter optimizations, the model was fitted to the experimental data of all temperatures simultaneously. Parameters b_0, $E_{\min}$, and $E_{\max}$ are assumed temperature-independent while the maximum adsorption concentration $C_{\mu s}$ is allowed to vary with temperature. The Unilan equation is in good agreement with experimental data for all three species, except some deviations exist for carbon dioxide at low-pressure region (< 3 kPa) and propane at low pressure (< 1 kPa). The model fits by a Toth equation are also shown in Figure 1 as solid-dashed lines, but at a temperature of 30°C only, since experimental data of one temperature is sufficient to extract the parameters in the Toth equation. The results from the Toth equation are similar to those from the Unilan equation and slightly better at the low-pressure region. The isotherm parameters for Unilan and Toth equations are tabulated in Tables 1 to 4 respectively. The Toth equation has the following form:

$$C_\mu = C_{\mu s} \frac{b^{1/t} p}{(1 + b p^t)^{1/t}} \qquad (5)$$

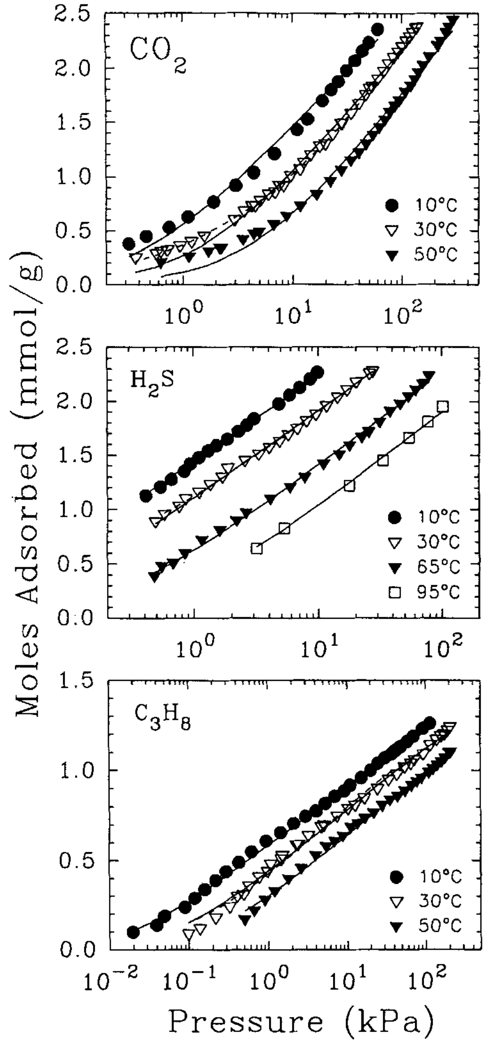

Figure 1. Adsorption equilibrium isotherm of gases on hydrogen mordenite.

(——) Unilan; (– – –) Toth.

and the Unilan equation is

$$C_\mu = \frac{C_{\mu s}}{2s}\ln\left(\frac{1+\bar{b}e^{s}p}{1+\bar{b}e^{-s}p}\right);\quad \bar{b} = b_0\exp\left(\frac{E_{\max}+E_{\min}}{2R_gT}\right);$$

$$s = \frac{E_{\max}-E_{\min}}{2R_gT} \tag{6}$$

Table 2. Langmuir-Uniform Energy Distribution Isotherm Parameters of Hydrogen Sulfide on Hydrogen Mordenite

Temp. °C	$C_{\mu s}$ mmol/g	b_0 kPa^{-1}	$E_{\min}$ J/mol	$E_{\max}$ J/mol
10	5.697	4.613×10^{-6}	0.42	38,720
30	5.466			
65	5.113			
95	4.824			

The extracted parameters were then used to predict the binary adsorption equilibria of carbon dioxide, hydrogen sulfide and propane in the same particle (hydrogen mordenite). Figure 2 shows the binary equilibrium mole fraction and total adsorbed amount of CO$_2$ and C$_3$H$_8$ at 30°C and 40.93 kPa. Azeotropic behavior is observed for these conditions. The predictions from five different models are compared with the experimental data. It is seen that the HIAST using a uniform energy distribution (solid lines) best predict the data, followed by HIAST-binomial distribution (dashed lines). Only these two models can predict the azeotropic behavior exhibited by CO$_2$-C$_3$H$_8$/H-mordenite system. It is interesting to note that Moon and Tien (1988) concluded that the HIAST could not produce the azeotropic system without extra site-matching parameters extracted from binary experimental data. In their study the same uniform energy distribution was also used. This unfortunate conclusion was made because proper procedures in determining isotherm parameter were not done. Due to computation difficulty Moon and Tien did not obtain the parameters with all parameters extracted simultaneously. Instead they obtained the maximum adsorption capacity from a Langmuir equation and then used that parameter as a constant to determine other isotherm parameters in the data fitting. Apparently, such a procedure cannot fit the single-component adsorption equilibrium data well (Figure 1 in Moon and Tien, 1988), especially at high-pressure range. The isotherm fitting at low pressure as well as high pressure will significantly affect the prediction of HIAST or IAST. This will be further discussed later. Therefore, the isotherm parameters obtained in Moon and Tien (1988) are not reliable in their use in multicomponent predictions. Furthermore, even the result of Moon and Tien could fit the binary adsorption equilibrium mole fraction well by adding the extra site-matching parameters; it could not reveal the maximum behavior of the total adsorbed amount. It is therefore emphasized here that any theory should first be in good agreement with the single-component experimental equilibrium data before it could be applied to predict multicomponent adsorption systems.

As seen in Figure 2, the HEL model is in significant error with the experimental data since the single-component maximum adsorption capacity of CO$_2$ (8.954 mmol/g) significantly

Table 1. Langmuir-Uniform Energy Distribution Isotherm Parameters of Carbon Dioxide on Hydrogen Mordenite

Temp. °C	$C_{\mu s}$ mmol/g	b_0 kPa^{-1}	$E_{\min}$ J/mol	$E_{\max}$ J/mol
10	8.553	1.537×10^{-8}	0.028	44,446
30	8.954			
50	9.130			

Table 3. Langmuir-Uniform Energy Distribution Isotherm Parameters of Propane on Hydrogen Mordenite

Temp. °C	$C_{\mu s}$ mmol/g	b_0 kPa^{-1}	$E_{\min}$ J/mol	$E_{\max}$ J/mol
9.9	1.494	1.208×10^{-6}	17,380	41,480
30	1.460			
51	1.409			

Table 4. Toth Isotherm Parameters of Gases on Hydrogen Mordenite at 30°C

Species	$C_{\mu s}$ (mmol/g)	b (kPa^{-t})	t
Carbon Dioxide	10.52	0.993	0.208
Hydrogen Sulfide	5.710	2.717	0.193
Propane	1.535	1.820	0.341

differs from that of C_3H_8 (1.460 mmol/g). Hence the use of extended Langmuir equation to describe the local adsorption equilibria is in serious error.

The IAST using either a Unilan or a Toth equation fails to predict the nonideality behavior represented by the azeotropic phenomenon. There is also some difference between the predictions of Toth and Unilan equations, although they equally describe the single-component adsorption isotherm fairly well.

The binary adsorption equilibria of H_2S and C_3H_8 at 30°C and 8.13 kPa are shown in Figure 3. None of the models predicts data well and/or can reveal the maximum in the total adsorbed amount. Nevertheless, among the five models the HIAST-uniform distribution is no doubt in best agreement with the experimental data. The HIAST-binomial is the second. The difference between the HEL and other models is narrowed in this case compared to the observation in Figure 2. This is probably due to the fact that the maximum adsorption capacities of H_2S (5.446 mmol/g) and C_3H_8 (1.460 mmol/g) are closer than those of CO_2 and C_3H_8. The global IAST models are again not as good as the HIAST, regardless what single-component isotherm equation is used. The IAST-Unilan model gives better predictions than the IAST-Toth equation for both mole fraction and total adsorbed amount.

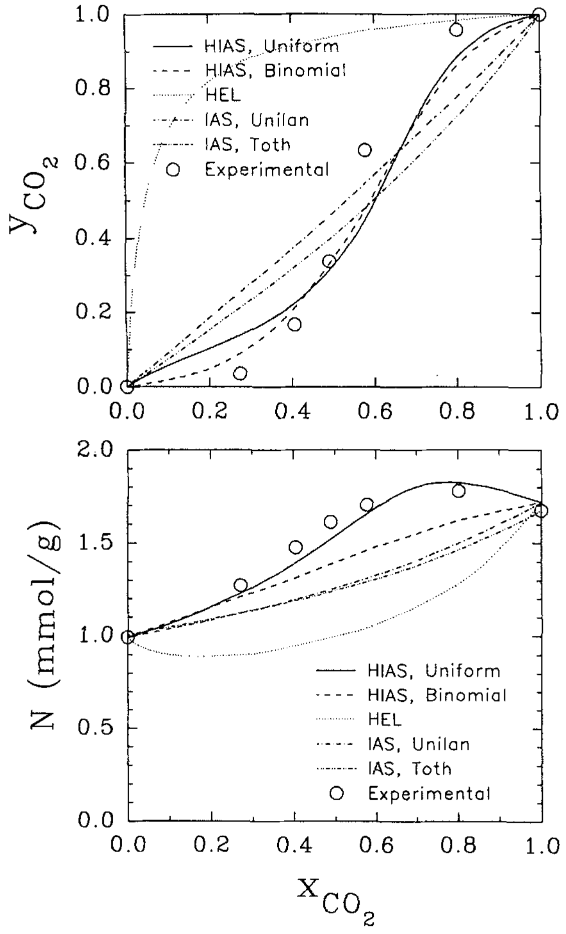

Figure 2. Prediction of binary adsorption of CO_2 and C_3H_8 on H-mordenite at 30°C and 40.93 kPa.

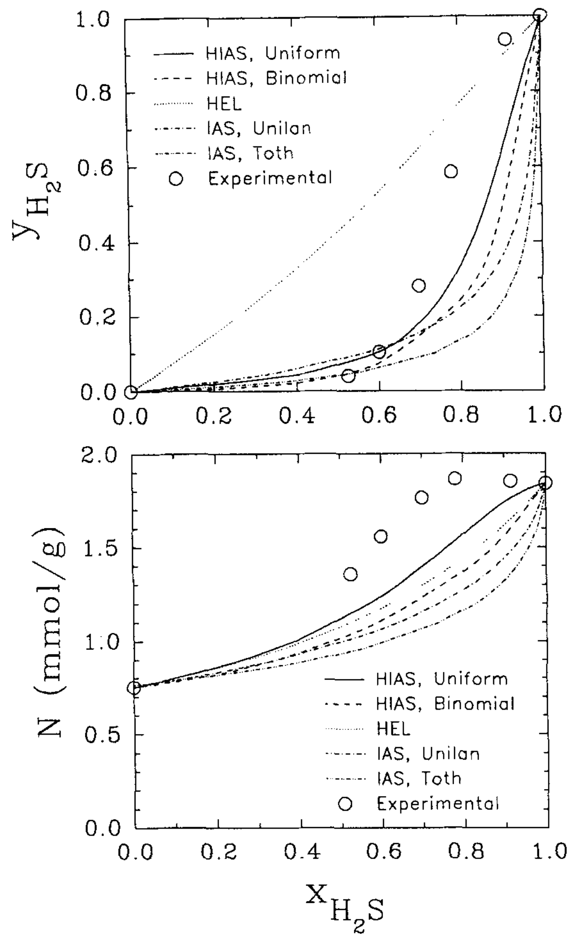

Figure 3. Prediction of binary adsorption of H_2S and C_3H_8 on H-mordenite at 30°C and 8.13 kPa.

Figure 4 shows the binary adsorption equilibria of H_2S and CO_2 at 30°C and 15.55 kPa. Since this system is quite ideal, all models except HEL predict the experimental data well. The IAST-Unilan is marginally better than IAST-Toth, and the same was observed in Figure 3 for the system of H_2S-C_3H_8/H-mordenite. We could conclude at this point that the HEL model should not be used in the multicomponent adsorption equilibria if the maximum adsorption capacity significantly differs from component to component. The HIAST is always better than the global IAST, and a continuous uniform energy distribution is better than a discrete binomial distribution when utilized in the HIAST.

To show the importance of the single-component isotherm fitting in the multicomponent prediction of HIAST, we chose

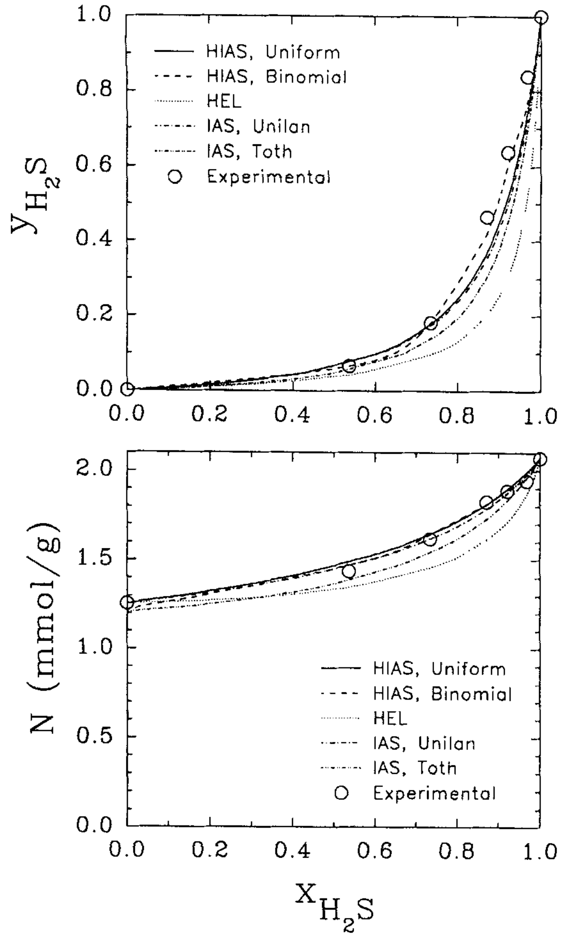

Figure 4. Prediction of binary adsorption of H_2S and CO_2 on H-mordenite at 30°C and 15.55 kPa.

the same Langmuir-uniform energy distribution (Unilan) equation, however, we fit the model to a different range of experimental data of CO_2 on H-mordenite. The first one was the same as plotted in Figure 1, that is, using the full pressure range in the fitting. In the second choice we fitted the isotherm equation to experimental data of less than 60 kPa. As expected, the second approach certainly has a better fit at the low-pressure region because of the narrow range, as seen in Figure 5 as dashed lines. However, when we extended the results of the second approach to the high-pressure region, the agreement between the model and the data is not good. As in Moon and Tien (1988), one may argue that the good fitting at the high-pressure region is not important in the multicomponent equilibrium prediction if the total pressure is low. Unfortunately, this is incorrect. In Figure 6 we compare the prediction results of the HIAST on the binary equilibria of CO_2 and C_3H_8 on H-mordenite using these two approaches of single-component isotherm fitting. To isolate the contribution of the choice of pressure range in the fitting of single-component isotherm, we used the full pressure range for fitting propane data and used the two approaches mentioned above for the fitting of carbon dioxide data. In the first approach, the full range was used in the parameter optimization and in the second only those CO_2 data of less than

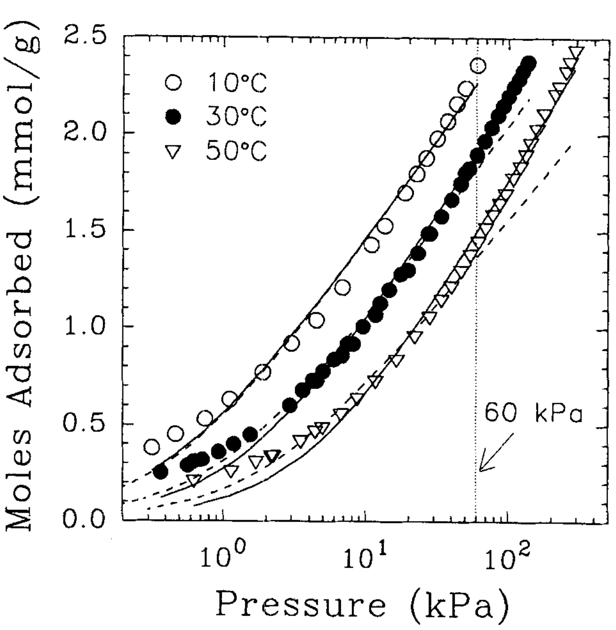

Figure 5. Adsorption equilibrium isotherm of CO_2 on hydrogen mordenite.

(——) fitting to full range; (– – –) fitting up to 60 kPa.

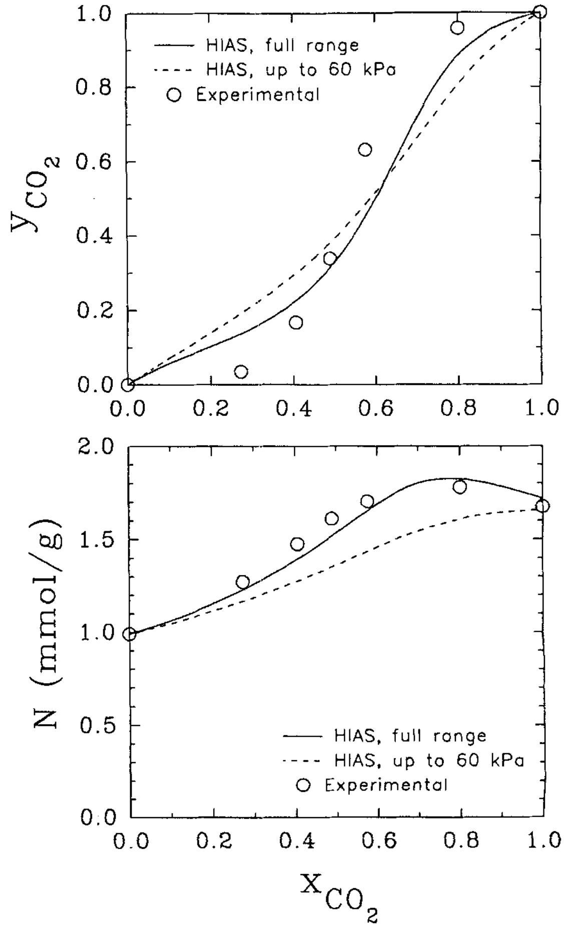

Figure 6. Prediction of binary adsorption of CO_2 and C_3H_8 on H-mordenite at 30°C and 40.93 kPa.

60 kPa was used in the isotherm parameters extraction (Figure 5). The results in Figure 6 obviously show that the second approach is significantly worse than the first one where the full range of experimental data were utilized in the determination of isotherm parameters. This observation might be explained as follows. Although the total pressure of the binary system (40.93 kPa) is less than the maximum pressure cut off (60 kPa) in the isotherm parameter optimization of the second approach, a higher value of pressure than the system total pressure may be required in the HIAST calculations of spreading pressure of pure component. Therefore, the model fit of single-component equilibrium at pressures higher than the binary total pressure will affect the prediction of HIAST. In short, isotherm fitting of pure component data must be carried over the full available experimental data.

In the previous study we concluded that the HEL is not suitable for the prediction of binary adsorption equilibrium because of the significant difference in the maximum adsorption capacities of different components. However, with special treatment in the single-component equilibrium data analysis forcing the maximum adsorption capacity to be the same for all species, the thermodynamic consistency is satisfied. If this is applied to the HEL model as well as the HIAST-uniform, then the approach of HEL is identical to HIAST-uniform. However, the pure component isotherm fit of the HEL is not as good as the original approach because of the requirement of maximum adsorbed capacities to be the same for all species. The question is simply whether the agreement between the model and data is still good enough after this restriction in the parameter optimization. The isotherm forced fitting of the maximum adsorption capacity to be the same for different species is shown in Figure 7 for CO_2, H_2S, and C_3H_8 on the H-mordenite at 10 and 30°C. Two different treatments were carried out. One found the necessary parameters using the full range of experimental data (solid lines), and the other obtained the parameters utilizing only those data less than 50 kPa (dashed lines). As seen in Figure 7, both methods can describe the experimental data well with the one using less data points giving a slightly better fit in the low-pressure region and a slightly worse extrapolation in the high-pressure end for carbon dioxide. The reason for the different parameter extraction methods is to further confirm the argument we made in the previous paragraph in the prediction of HIAST, which is affected by the treatment of the single-component equilibrium data. However, in the prediction of HEL on the binary equilibria it is expected that a good agreement between the model and only those data less than the total pressure of multicomponent systems is adequate, because in the multicomponent equilibrium calculation using HEL no information of the pure component data above the total pressure is required. This is confirmed by the prediction result of binary CO_2 and H_2S on H-mordenite shown in Figure 8. The predictions of the *x-y* diagram at constant pressure plane by HEL are superimposed to each other using the single-component isotherm parameters extracted either from full data range or up to 50 kPa only. Between them there is some small difference on the total adsorbed amount prediction on the CO_2 side, which is caused by the different fitting results on the single-component isotherm of CO_2. Another interesting observation is that the HEL can predict the azeotropic behavior of the binary system when the maximum adsorption capacity is forced to be the same for different species, which is a significant improvement over the original HEL prediction (Figure 2). The HEL prediction on multicomponent adsorption equilibria can be therefore improved and better than global IAST by forcing different species to have the same maximum adsorption capacity in the single-component isotherm parameter extractions. Although this result is not as good as the HIAST, it saves the computation time as the equations are explicit and requires no iteration.

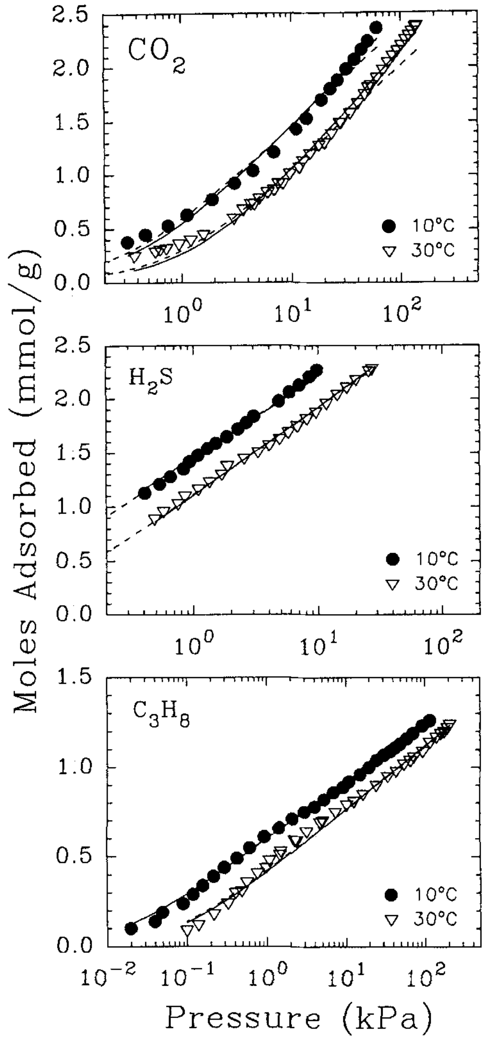

Figure 7. Adsorption equilibrium isotherm of gases on hydrogen mordenite.

(——) fitting to full range; (– – –) fitting up to 50 kPa.

We now turn to ternary systems to test the prediction capacity of the five models. A percent deviation parameter on total adsorbed amount D was utilized here to compare the prediction results of different methods:

$$D = \left| \frac{N_{\text{exp}} - N_{\text{pre}}}{N_{\text{exp}}} \right| \times 100 \tag{6}$$

where N is the total adsorbed amount and the results are listed in Table 5 together with those of binary systems. The

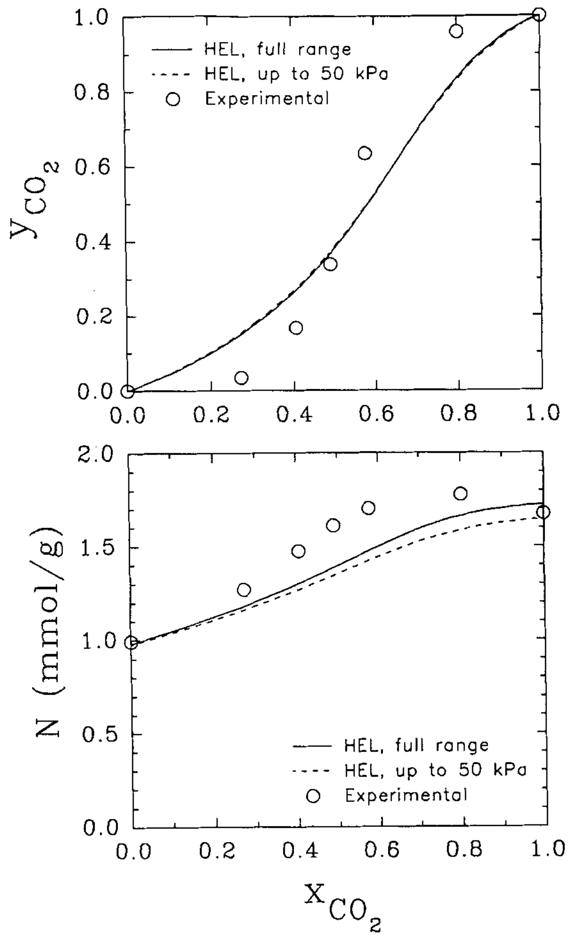

Figure 8. Prediction of binary adsorption of CO_2 and C_3H_8 on H-mordenite at 30°C and 40.93 kPa.

HIAST-uniform energy distribution, is in best agreement with the experimental data, followed by HEL with same $C_{\mu s}$ for different components, IAST-Toth and IAST-Unilan. The HEL with different $C_{\mu s}$ is the worst.

Conclusions

Applying IAST to a local site and then doing the integration over the full energy distribution (HIAST) is a better approach in predicting multicomponent adsorption equilibria than the treatment of global IAST. By attributing the nonideality to the surface heterogeneity, the HIAST is a recommended model for predictions describing well the azeotropic phenomenon of a binary adsorption system. When used in the HIAST to predict the multicomponent adsorption equilibria, a continuous uniform energy distribution gives a different result from a discrete binomial distribution. It should be careful in concluding the goodness of the HIAST in representing experimental data since its prediction on multicomponent equilibria depends on the choice of energy distribution and the appropriate determination of the single-component isotherm parameters. By properly choosing a functional form describing the energy distribution, the predictions of HIAST could be improved. The usage of HEL equation is not encouraged unless the maximum adsorption capacities of all species considered are the same or very close to each other. The global IAST prediction also depends on the choice of the single-component isotherm equation.

Table 5. Deviations on the Total Adsorbed Amount Prediction of Multicomponent Equilibria

	D(%)					
System	HIAS-Uniform	IAS-Unilan	IAS-Toth	HEL	HEL (Same $C_{\mu s}$)	No. Point
$CO_2+C_3H_8$	13	23	22	35	17	9
$H_2S+C_3H_8$	20	27	25	38	22	8
CO_2+H_2S	6	6	3	6	7	9
$CO_2+H_2S+C_3H_8$	11	17	16	35	12	10

Acknowledgments

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Notation

b = Toth isotherm parameter, kPa^{-t}
$\bar{b}$ = Unilan isotherm parameter, kPa^{-1}
b_0 = affinity constant at zero energy level, kPa^{-1}
C_μ = adsorbed phase concentration, mol/g
$C_{\mu s}$ = maximum adsorbed capacity, mol/g
D = deviation parameter
E = adsorbate-adsorbent interaction energy, J/mol
F = energy distribution function
N = total adsorbed amount, mol/g
p = gas-phase concentration, kPa
R_g = gas constant, J/(mol·K)
s = Unilan isotherm parameter
t = Toth isotherm parameter
x = mole fraction in the adsorbed phase
y = mole fraction in the gas phase

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