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Multisite Langmuir Model Applied to the Interpretation of Sorption of *n*-Paraffins in 5A Zeolite

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Adsorption equilibrium isotherms of *n*-paraffins from methane to decane on 5A zeolites (crystals and pellets) were compiled in a wide range of temperature and pressure. The data were correlated using the MSL (multisite Langmuir) model for homogeneous surfaces. It is shown that it is possible to correlate all adsorption isotherms by maintaining thermodynamic consistency on model parameters. However, for paraffins with more than seven carbons it is necessary to account for the effects of lateral interaction of adsorbed molecules. The heat of adsorption for C₁–C₁₀ paraffins linearly increases with the carbon number, ranging from 4.6 kcal/mol for methane up to 23.2 kcal/mol for *n*-decane.

Introduction

The equilibrium of adsorption is important information needed for the design of an adsorber. A correct interpretation and quantification of adsorption equilibrium isotherms is required in the development of adsorption technologies for the separation and purification of gases and liquids. Langmuir¹ considered that the adsorbent is constituted by several active sites that can accommodate only one molecule, or if the molecule dissociates when it adsorbs, each atom occupies one active site. This is called localized adsorption with no interaction between adsorbed molecules. Langmuir derived the following isotherm based on the dynamic equilibrium between adsorbed and gas-phase species assuming that a molecule when it adsorbs does not dissociate:

$$K_{\text{eq}} = \frac{1}{p} \frac{\theta}{1 - \theta} \quad (1)$$

where the equilibrium constant $K_{\text{eq}} = k_{\infty} \exp(-\Delta H_{\text{ads}}/RT)$. The Langmuir equation has only one parameter in terms of correlation of experimental data $\theta = q/q_{\text{max}}$ versus p because q_{max} should be temperature independent and can be measured independently of the shape of the isotherm. Several experimental results have been described by this equation, letting the parameter q_{max} change with temperature. A correlation of this type does not give physical meaning to the model being fit purely empirically. However, the Langmuir isotherm is the most used isotherm to interpret isotherms of type I.

When the physical validity of the Langmuir isotherm is to be maintained, generally the experimental isotherm is not well described by the model. Several corrections of the model have been proposed, namely, the isotherm of Fowler and Guggenheim,²

$$K_{\text{eq}} \exp\left(\frac{2u\theta}{RT}\right) = \frac{1}{p} \frac{\theta}{1 - \theta} \quad (2)$$

which takes into account possible interactions between adsorbed molecules through the parameter u , which is negative for repulsion and positive for attraction.

More recently, Nitta et al.,³ using arguments of statistical thermodynamics and assuming that a molecule when it adsorbs can occupy n active sites, developed the multisite Langmuir (MSL) isotherm

$$K_{\text{eq}} \exp\left(\frac{nu\theta}{kT}\right) = \frac{1}{p} \frac{\theta}{(1 - \theta)^n} \quad (3)$$

where u is the interaction energy between adsorbed molecules (positive for attraction and negative for repulsion) and k is the Boltzmann constant. Sircar⁴ derived the above isotherm, using kinetic arguments of the Langmuir type, neglecting the interaction term between adsorbed molecules ($u = 0$); he also showed that MSL mixed-gas results (thermodynamically consistent), such as prediction of azeotropes, cannot be explained by ideal adsorption solution theory (IAST) of Myers and Prausnitz⁵ because IAST does not explicitly cover the effect of adsorbate sizes.

A more complete isotherm taking into account the effects of adsorbate size, chemical dissociation, and molecular interactions has been recently proposed by Martinez and Basmadjian,⁶ i.e.,

$$K_{\text{eq}} \exp\left(\frac{nu\theta}{kT}\right) = \frac{1}{p} \frac{\theta^s}{(1 - \theta)^n} \quad (4)$$

where s represents the dissociation parameter. This isotherm reduces to the Langmuir one when there is no dissociation ($s = 1$), there are no interactions ($u = 0$), and a molecule occupies only one active site when it adsorbs ($n = 1$).

To maintain thermodynamic consistency, Nitta et al. and Martinez and Basmadjian models require that the relation

$$nq_{\text{max}}/M_w = \text{constant} \quad (5)$$

be obeyed.

In this paper emphasis is given to the analysis of available adsorption equilibrium data of *n*-paraffins in 5A zeolites using a localized adsorption model (MSL model) developed by Nitta et al.³ In previous works it has been shown by Silva and Rodrigues^{7,8} that the MSL model represents with good accuracy the adsorption equilibrium of *n*-pentane and *n*-hexane in 5A zeolite

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Table 1. MSL Model Parameters for the Sorption of *n*-Paraffins in 5A Zeolite Pellets^a

<i>n</i> -alkane	adsorbent	temp (K)	original model		MSL model					
			Henry's constant <i>H</i> (g/g _{ads} ·bar)	heat of adsorption −Δ <i>H</i> _{ads} (kcal/mol)	Henry's constant <i>H</i> = <i>K</i> _{eq} <i>q</i> _{max} (g/g _{ads} ·bar)	heat of adsorption −Δ <i>H</i> _{ads} (kcal/mol)	<i>n</i>	interaction parameter <i>u/k</i> (K)	<i>q</i> _{max} (g/100 g _{ads})	DN _{av} ^{<i>b</i>}
methane	crystals ^{<i>c</i>}	195		5.2	0.833	4.6	1.4		10.3	0.27
		212	0.454	5.2	0.321	4.6	1.4		10.3	0.10
		230	0.180	5.2	0.137	4.6	1.4		10.3	0.13
		253	0.060	5.2	0.055	4.6	1.4		10.3	0.03
ethane	crystals ^{<i>d,e</i>}	230 ^{<i>d</i>}	17.5	6.9	8.59	6.0	2.4		11.3	0.70
		273 ^{<i>d</i>}	1.17	6.9	1.09	6.0	2.4		11.3	0.25
		308 ^{<i>e</i>}		6.9	0.31	6.0	2.4		11.3	0.11
		324 ^{<i>d</i>}	0.24	6.9	0.19	6.0	2.4		11.3	0.12
		345 ^{<i>d</i>}	0.095	6.9	0.11	6.0	2.4		11.3	0.06
		323			4.81	8.1	3		13	0.23
propane	pellets ^{<i>f</i>}	348			1.95	8.1	3		13	0.11
		323			91.0	10.2	4		13	0.27
		358			19.2	10.2	4		13	0.10
<i>n</i> -butane	pellets ^{<i>g</i>}	398			4.55	10.2	4		13	0.27
		498			0.342	10.2	4		13	0.18
		373			144	13.2	5		13	0.07
		423			16	13.2	5		13	0.11
<i>n</i> -pentane	pellets ^{<i>h</i>}	473			3.2	13.2	5		13	0.09
		523			0.82	13.2	5		13	0.15
		573			0.28	13.2	5		13	0.08
		373			1436	14.2	6		13	0.03
		523			152	14.2	6		13	0.03
<i>n</i> -hexane	pellets ^{<i>i</i>}	473			26	14.2	6		13	0.03
		523			6.76	14.2	6		13	0.04
		573			1.66	14.2	6		13	0.06
		439 ^{<i>j</i>}	253	16.5	228	16.9	7	379	13	0.29
		462 ^{<i>j</i>}	94.1	16.5	86.6	16.9	7	379	13	0.17
		491 ^{<i>j</i>}	33.8	16.5	29.0	16.9	7	379	13	0.15
<i>n</i> -heptane	crystals ^{<i>j</i>} + pellets ^{<i>k</i>}	573 ^{<i>k</i>}		16.5	2.43	16.9	7	379	13	0.25
		523	50.7	19.2	39.0	19.2	8	392	13	0.14
		568	9.9	19.2	9.02	19.2	8	392	13	0.10
		617	2.9	19.2	2.31	19.2	8	392	13	0.06
<i>n</i> -octane	crystals ^{<i>l</i>}	668	0.82	19.2	0.707	19.2	8	392	13	0.04
		523	407	23.2	390	23.2	10	392	13	0.28
		545	162	23.2	158	23.2	10	392	13	0.12
		573	62.2	23.2	55.5	23.2	10	392	13	0.22
<i>n</i> -decane	crystals ^{<i>l</i>}	596	23.2	23.2	25.3	23.2	10	392	13	0.07

^a All parameters shown above are referred to adsorption in pellets. To convert the adsorbed amount from molecules/cavity in the crystals to g/g_{ads} in the pellets, the relation 1 molecule/cavity = 0.45 mmol/g_{pellet} was used (Ruthven, 1984). ^b DN_{av} is defined as the average of the absolute values of the difference between experimental and calculated results (Valenzuela and Myers, 1989). ^c Ruthven (1976). ^d Ruthven and Loughlin (1971). ^e Glessner and Myers (1969). ^f Ruthven and Derrah (1972). ^g Ruthven and Loughlin (1971). ^h Silva and Rodrigues (1997a). ⁱ Silva and Rodrigues (1997b). ^j Doetsch et al. (1974). ^k Jullian (1993). ^l Vavlitis et al. (1981).

pellets. The aim of the work is to use the MSL model to interpret the sorption data of *n*-paraffins from methane to decane in 5A zeolites (crystals and pellets) using data published in the literature from various authors and our own laboratory.

Study of the Sorption of Normal Paraffins in 5A Zeolite

To study the validity of the MSL model relative to the adsorption of *n*-paraffins in 5A zeolites, experimental data published in the literature from methane to *n*-decane were compiled by Silva⁹ and reported in Table 1. The isotherms of methane, ethane, *n*-heptane, *n*-octane, and *n*-decane obtained with 5A zeolite crystals were reported in molecules/cavity as a function of pressure; to express the amount adsorbed in g/g_{ads}, the relation 1 molecule/cavity = 0.45 mmol/g of Linde pellet was used.¹⁰ It should be noted that the amount adsorbed in g/g_{ads} of crystals is different from the amount adsorbed in g/g_{ads} of pellets because of the addition of inert clay binder to crystals necessary to form the pellets. Experimental results for adsorption isotherms of ethane, propane, *n*-butane, *n*-pentane, and *n*-hexane in pellets are also available; the amount adsorbed is given directly in mmol/g_{pellet} or g/100 g_{pellet}. As noted previously, to

maintain thermodynamic consistency of the MSL model, the relation $nq_{\max}/M_w = \text{constant}$ should be obeyed. The maximum adsorbent capacity, $q_{\max} = 13 \text{ g/100 g}_{\text{ads}}$, was fixed during the fitting of *n*-pentane and *n*-hexane isotherms in 5A zeolite pellets obtained in our laboratory. The data of Pal et al.¹¹ for adsorption of *n*-heptane and *n*-dodecane in 5A zeolite also show values on the order of 15 g/100 g_{ads} for the maximum adsorbed capacity in zeolite crystals.

Isotherms of Methane, Ethane, and Propane in 5A Zeolite. Data for methane¹² and ethane^{13,14} sorption in crystals and propane in 5A Davison C-521 pellets¹⁵ are shown in Figure 1a–c, respectively. Experimental data for methane and ethane were originally fitted with the statistical model.¹⁶ The heat of adsorption is around 5.2 kcal/mol for methane and 6.9 kcal/mol for ethane.

The MSL model was used to fit the data for all adsorbates with the interaction parameter *u* set equal to zero. The values of *K*_{eq}, *n*, and *q*_{max} are shown in Table 1. The isosteric heat of adsorption is 4.6 kcal/mol for methane, 6.9 kcal/mol for ethane, and 8.1 kcal/mol for propane, similar to that reported in the literature by Vavlitis et al.¹⁷

The values of the parameters *n* and *q*_{max} of the MSL model are 1.4 and 10.3 g/100 g_{ads}, respectively, for meth-

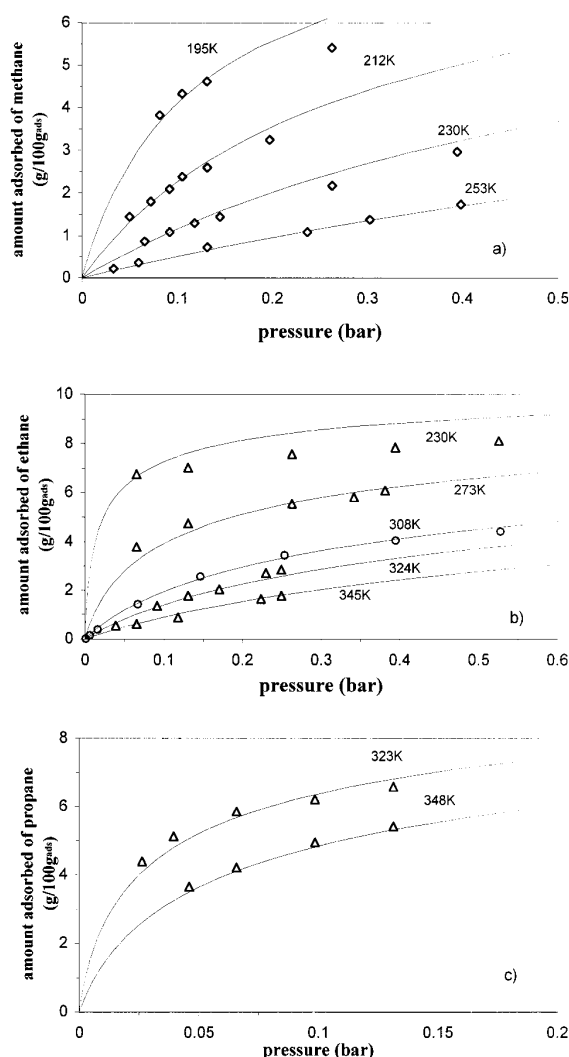


Figure 1. Adsorption equilibrium isotherms of methane, ethane, and propane in 5A zeolites: (a) methane (data of Ruthven¹² for crystals); (b) ethane (data of Ruthven and Loughlin¹³ (Δ) and Glessner and Myers¹⁴ (\circ) for crystals); (c) propane (data of Ruthven and Derrah¹⁵). Points are experimental data. Lines are MSL model results. Absolute temperatures are quoted in each curve.

ane, 2.4 and 11.3 g/100 g_{ads} for ethane, and 3 and 13 g/100 g_{ads} for propane. The maximum adsorbent capacity of the sample used in the sorption of methane and ethane is lower than 13 g/100 g_{ads}; it was not possible from the data shown in the work of Ruthven to calculate the maximum adsorbent capacity in order to assess the validity of the value selected. The MSL model isotherm predicts with accuracy the isotherms between 273 and 345 K but fails at the temperature of 230 K.

Isotherms of *n*-Butane, *n*-Pentane, and *n*-Hexane on 5A Zeolite. Parts a–c of Figure 2 show data for *n*-butane sorption on Linde 5A molecular sieve pellets¹⁸ and *n*-pentane and *n*-hexane in 5A zeolite pellets from Rhone-Poulenc.^{7,8} The data for *n*-butane were originally fitted by the Dubinin–Polanyi potential theory by use of a characteristic curve.

The MSL model was fitted to all isotherms using the same $q_{\max} = 13$ g/100 g_{ads} and n equal to 4, 5, and 6 for *n*-butane, *n*-pentane, and *n*-hexane, respectively. The isosteric heats of adsorption are 10.2 kcal/mol for

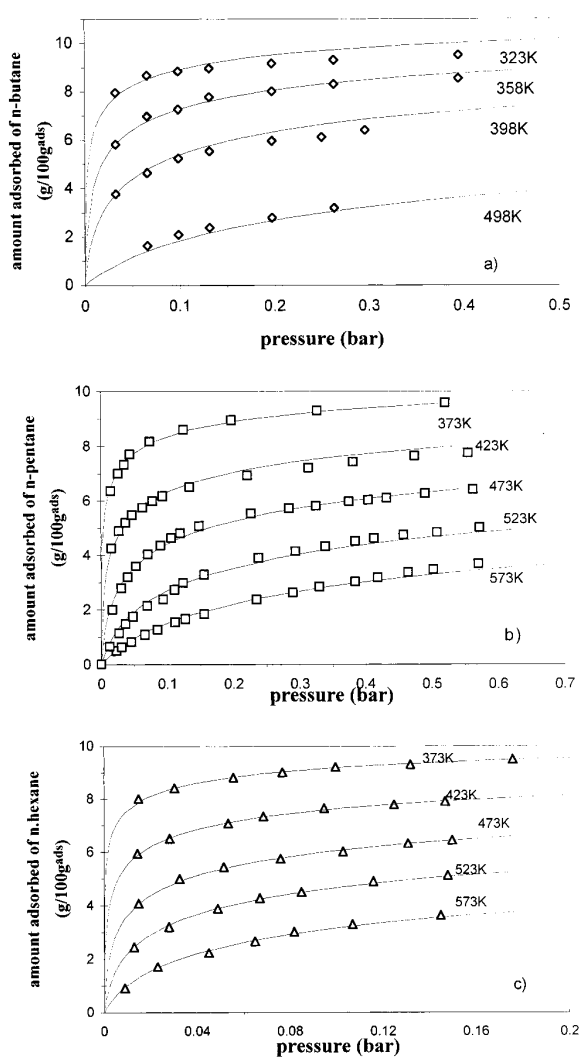


Figure 2. Adsorption equilibrium isotherms of *n*-butane, *n*-pentane, and *n*-hexane in 5A zeolite: (a) *n*-butane (data of Ruthven and Loughlin¹⁸); (b) *n*-pentane (data from Silva and Rodrigues⁷); (c) *n*-hexane (data from Silva and Rodrigues⁸). Points are experimental results. Lines are MSL model results. Absolute temperatures are quoted in each curve.

n-butane, in good agreement with the value published by Vaylitis et al.,¹⁷ 13.2 kcal/mol for *n*-pentane, and 14.2 kcal/mol for *n*-hexane. Table 1 summarizes the parameters.

Isotherms of *n*-Heptane, *n*-Octane, and *n*-Decane in 5A Zeolite. The sorption of *n*-heptane in 5A zeolite crystals was studied by Doetsch et al.¹⁹ in a gravimetric system. Original sorption data in molecules/cavity are plotted in logarithmic scales, and it is difficult to recover the experimental points. The strategy followed was to plot the model isotherms used by Doetsch et al. to fit their experimental data with the model parameters shown in their work. Figure 3a shows such a representation. The experimental data of Doetsch et al. were fitted originally by two completely different models: (i) one assumes that the sorption occurs in a system of independent pairs of equivalent sites with an interaction energy when both sites of a pair are occupied; (ii) the other assumes sorption with negligible interaction between adsorbed molecules on a set of

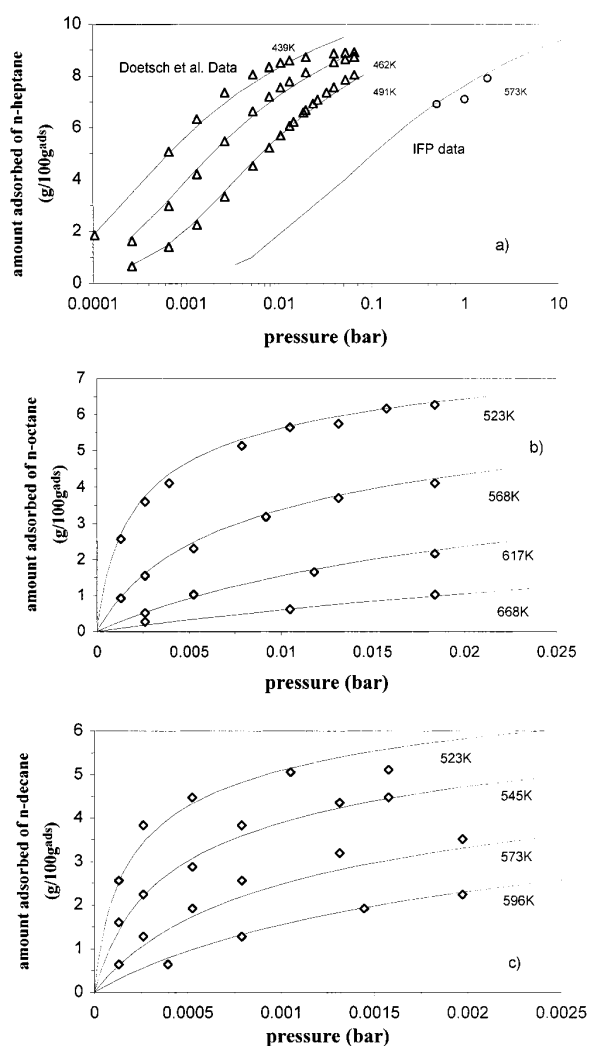


Figure 3. Adsorption equilibrium isotherms of *n*-heptane, *n*-octane, and *n*-decane in 5A zeolite: (a) *n*-heptane (data of Doetsch et al.¹⁹ and IFP of Jullian²⁰); (b) *n*-octane in 5A zeolite (data of Vavlitis et al.¹⁷); (c) *n*-decane in 5A zeolite (data of Vavlitis et al.¹⁷). Points are experimental data. Lines are MSL model results. Absolute temperatures are quoted in each curve.

independent pairs of nonequivalent sites. The two models predict with good accuracy all adsorbed behavior. The evidence from experimental data of the precise saturation limit of 2 molecules/cavity and the lack of variation of this limit with temperature is the basis for the two model approximations.

For the fitting procedure of experimental data of Doetsch et al. with the MSL model shown in Figure 3a, $q_{\max} = 13 \text{ g/100 g}_{\text{ads}}$ and $n = 7$. It was found during the fitting procedure that to keep the consistency of parameter n , it is necessary to introduce the interaction parameter u/k to the MSL model according to eq 3. The value of u/k is 379 K, suggesting an attractive relation between sorbed molecules. This value is comparable with the force constant ($\epsilon/k = 399.3 \text{ K}$) for *n*-hexane. The attractive nature between sorbed molecules seems to be controversial; according to the statistical model of Ruthven, there is no need to account for the intermolecular attraction between adsorbed molecules in non-polar sorbates; only the attractive potential of the framework which determines Henry's constant and the

repulsive interaction between molecules due to their finite size are of importance. However, most of the localized adsorption models derived from statistical thermodynamics account for the attractive nature of adsorbed molecules, and significant data were fitted using the attractive parameter.³ An explanation for the need of introducing the parameter u in the fitting procedure is the increasing length of *n*-paraffins. After *n*-heptane, there is no space in the cage of 5A zeolite for more than two molecules; therefore, the conformation of sorbed molecules in the 5A zeolite cage leads to an attraction between sorbed molecules.

In Figure 3a the data of Jullian from IFP²⁰ for the sorption of *n*-heptane in 5A zeolite pellets at a pressure of near 2 bar and a temperature of 573 K are also plotted. It is remarkable that with the parameters obtained by the fitting procedure of the MSL model to the data of Doetsch et al. we were able to predict the sorption data of *n*-heptane at such high pressure and temperature. It seems that the MSL model is also effective for the correlation of data on sorption of *n*-paraffins in 5A zeolite. Table 1 summarizes the model parameters. The values of Henry's constant obtained by the MSL model between 439 and 573 K are very similar to those obtained from the virial isotherm according to the treatment of Doetsch et al. The values of heat of adsorption at zero coverage, obtained from the virial isotherm and the MSL model, are 16.5 and 16.9 kcal/mol, respectively.

Figure 3b shows experimental data for the sorption of *n*-octane in 5A zeolite crystals obtained by Vavlitis et al.¹⁷ between 523 and 668 K and for pressures up to 14 Torr. The author only extracts Henry's constants from the data because the main objective of their work was to study sorption kinetics. The MSL model fitting of data, keeping the thermodynamic consistency of the parameters, used $n = 8$ and $q_{\max} = 13 \text{ g/100 g}_{\text{ads}}$. Again it was necessary to introduce the parameter u/k in the fitting procedure. The value shown in Table 1 is 392 K, suggesting a strong attraction between sorbed molecules. The isosteric heats of adsorption summarized in Table 1 obtained by Vavlitis et al. (1981) and by the MSL model are 19.2 kcal/mol.

Figure 3c shows experimental data for the sorption of *n*-decane in 5A zeolite crystals obtained by Vavlitis et al.¹⁷ at temperatures between 523 and 668 K and pressures up to 1.6 Torr. The fitting with the MSL model used $n = 10$ and $q_{\max} = 13 \text{ g/100 g}_{\text{ads}}$. Again the parameter u/k is introduced; it is 392 K, which is equal to the value found in *n*-octane isotherms. The isosteric heat of adsorption is 23.2 kcal/mol. In all cases the quality of the fitting is given by DN_{av} , defined in Table 1 as in Valenzuela and Myers.²¹

Conclusions

The MSL model is effective for the correlation of the sorption of linear paraffins from C_1 to C_{10} in 5A zeolites (crystals and pellets). It is possible, by keeping the thermodynamic consistency of the parameters, to correlate data of sorption of methane at temperatures of near 195 K to sorption of *n*-decane at temperatures of near 668 K. The model indicates the increasing heat of adsorption with chain length, as shown in Figure 4. The heats of adsorption range from 4.5 kcal/mol for methane up to 23.2 kcal/mol for *n*-decane. In Figure 5 the values of Henry's constants plotted as a function of the reciprocal of absolute temperature are shown. The trends shown in Figure 5 are consistent: Henry's constants increase as the temperature diminishes; as the paraffins

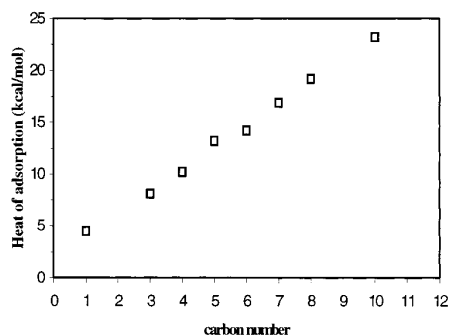


Figure 4. Isosteric heat of adsorption as a function of carbon number predicted by the MSL model for the sorption of *n*-paraffins in 5A zeolite pellets.

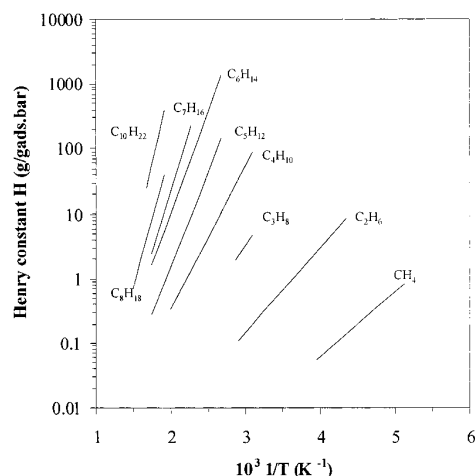


Figure 5. Temperature dependence of Henry's constants predicted by the MSL model.

increase in carbon number, the slope of the straight lines increases, indicating an increase in the heat of adsorption. Another interesting aspect of the plot is that for the paraffins with higher carbon number Henry's constants are not so different from those in the case of the light paraffins. Because Henry's constant is a measure of the effects of sorbate-sorbent attractions, it seems that for the hydrocarbons with higher carbon number these effects tend to be similar, which is not the case for the light paraffins. It was also found from the model that for paraffins with carbon number higher than C_7 there is a strong attraction between sorbed molecules. An explanation is that the space in the 5A cage is not enough for more than two molecules, and only by attractive forces does the adsorption content increase.

Nomenclature

ΔH_{ads} = heat of adsorption (kcal/mol)
 H = Henry's law constant (g/g \cdot bar)
 k = Boltzmann constant (J/K)
 k_{∞} = preexponential factor (g/g $_{ads}$ \cdot bar)
 K_{eq} = equilibrium constant (bar $^{-1}$)
 M_w = molecular mass of sorbate (g/mol)
 n = number of active sites occupied by a molecule
 p = partial pressure of sorbate (bar)
 q = amount adsorbed (g/100 g $_{ads}$)
 q_{max} = maximum amount adsorbed (g/100 g $_{ads}$)
 R = ideal gas law constant (J/mol \cdot K)

s = dissociation parameter

T = temperature in the bulk gas phase (K)

u = interaction energy (J)

Greek Letters

ϵ = energy constant for Lennard-Jones potential (J)

θ = fractional coverage of adsorbent ($=q/q_{max}$)

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