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Equilibrium Adsorption of Light Alkanes in Silicalite-1 by the Inertial Microbalance Technique

W. ZHU,* F. KAPTEIJN AND J.A. MOULIJN
Industrial Catalysis, DelftChemTech, Delft University of Technology Julianalaan 136,
2628 BL Delft, The Netherlands
w.zhu@tnw.tudelft.nl

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Abstract. The equilibrium adsorption of the light alkanes methane, ethane, propane, *n*-butane, and i-butane in silicalite-1 has been investigated using the TEOM technique. Either a conventional or a dual-site Langmuir isotherm appropriately describes the equilibrium data. Good agreement with the literature data determined by other techniques indicates the TEOM is a reliable technique. The adsorption of i-butane in silicalite-1 reveals the discrete preferential molecular siting, implying a discrete-dual-structural heterogeneity for light alkanes in silicalite-1.

Keywords: silicalite-1, adsorption, TEOM, alkanes, isosteric heat, adsorption entropy

Introduction

Adsorption data of components are of utmost importance for a good design of separation units based on differences in thermodynamics or in kinetic properties. Often data are obtained by either a volumetric method or a gravimetric method. Both methods have their advantages and disadvantages, and generally large corrections have to be made on the raw experimental data.

Recently, a new method has been commercialized in which the mass change of a vibrating tube is determined by frequency measurements, the so-called tapered element oscillating microbalance (TEOM) mass analyzer. The main feature of the TEOM technique is an oscillating element that is based on inertial forces, instead of weight, to measure the amount adsorbed. An important advantage of this technique is that the gas which has to adsorb flows through a packed bed of adsorbent material. So, external mass transport limitations are nearly absent in this technique. In addition, it is easy to maintain essentially isothermal conditions. Therefore, the TEOM can be used not only for steady state measurements, but also for transient uptake and desorption experiments.

This paper discusses the measurements of single component adsorption isotherms in silicalite-1 crystals using the TEOM technique. An interpretation of the difference between the isotherm of i-butane and the isotherms of the normal alkanes is given. The conventional and the dual-site Langmuir isotherm models have been used to describe the adsorption data. In addition, thermodynamic properties, like the isosteric heat and entropy of adsorption calculated from the isotherms, are presented to characterize interactions between adsorbate and adsorbent.

Experimental

A Rupprecht & Patashnick TEOM 1500 pulse mass analyser (100 mg sample volume) was used in an experimental set-up designed for measurement of equilibrium adsorption in microporous materials such as zeolites. The active element of the TEOM consists of a tube constructed of a material having a special taper. A feed back system maintains the oscillation of

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*Author to whom correspondence should be addressed.

the tapered tube. The natural frequency will change in relation to the mass in the tapered tube. The mass uptake is then determined by the change of the oscillating frequency. Software and hardware provided by R&P perform the required frequency counting and mass calculation. A LabView program is used to operate the set-up and to record the results. A detailed description of the TEOM apparatus is available in the literature (Chen et al., 1996).

Based on the operating principle of the TEOM, the total mass uptake consists of the amount adsorbed and the mass change caused by the change of the gas density in the tapered tube. If the TEOM is oscillating at the start of the experiment with the frequency of f_0 and exhibits the frequency f_1 after a mass uptake, the total mass uptake can be obtained as a function of f_0, f_1, and the spring constant K_0.

$$\Delta M = \Delta M_\mathrm{a} + \Delta M_\mathrm{g} = K_0\left(\frac{1}{f_1^2} - \frac{1}{f_0^2}\right) \quad (1)$$

Where ΔM_a is the adsorbed mass and ΔM_g is the change in the gas density determined by measuring a mass change during a reference run under the same conditions. From Eq. (1) it follows that a mass change is measured with respect to the conditions at the beginning of the experiment. At the start of every experiment the mass is set to zero and this corresponds with a frequency f_0. This frequency depends slightly on the amount of sample in the tapered element and is in the order of 70 Hz.

The change in the gas density depends on the type of gas and the operating conditions. To correct for the mass change caused by the change in the density of the gas phase reference experiments have been performed. In a reference experiment the response of the TEOM is measured without adsorption taking place. Three different reference experiments were performed with 1) 55 mg quartz wool; 2) 55 mg quartz wool with 43 mg uncalcined silicalite-1; and 3) no sample. No significant difference in the mass change was observed among these three reference runs. In addition, the relationship between the mass change in the reference runs and the partial pressure of the adsorbing gas is almost linear. No exact correlation, however, was found for different gases to be able to rely on a kind of master curve for this correction.

The spring constant K_0 is a weak function of the temperature. The change in the spring constant with temperature over the range of 303 to 525 K corresponds with a maximum mass change of 1×10^{-5} gram. For the series of isotherms this means that an error of less than 1% is introduced by going from the lowest temperature (303 K) to the highest temperature (525 K).

The tapered element was loaded with 47 mg of silicalite-1 crystals. Quartz wool was used at the top and the bottom of the sample bed to keep the adsorbent particles firmly packed, which is essential for a stable measurement.

With the TEOM both pressure and temperature programmed runs can be performed. The isotherms were obtained by a step-wise increase of the partial pressure of the feed gas at fixed temperatures. The isobaric experiments were performed by temperature programmed runs at a fixed pressure. The partial pressure of the feed gas was determined by its fraction of the total molar-feed-flow rate through the sample bed and the total pressure. A mixture of helium and the adsorbate gas was used to create partial pressures below 1.013×10^5 Pa, and pure gas was used for pressures above 1.013×10^5 Pa.

When the partial pressure of the gas phase was changed, it took about 1 to 5 minutes to reach steady state. If the mass did not change for more than 1% in 2 minutes, equilibrium was assumed. This procedure was repeated and about every 10 minutes a new partial pressure could be set for most conditions. The amount adsorbed was then calculated by subtracting the reference run from the results obtained with the silicalite-1 crystals in the sample holder.

The silicalite-1 crystals with a ratio Si/Al > 1300 were supplied by Dr. J.C. Jansen (TU Delft). The template inside the silicalite-1 crystals was removed by calcination at 873 K for 12 hours. The apparent density of the sample was $1.76\ \mathrm{g \cdot cm^{-3}}$ and the adsorption of N_2 indicated an accessible microporous void volume of $0.179\ \mathrm{cm \cdot g^{-1}}$ and a microporous area of $372\ \mathrm{m^2 \cdot g^{-1}}$. The crystal size was about $120 \times 50 \times 50\ \mu$m $(L \times H \times W)$, as determined by SEM, shown in Fig. 1.

Prior to the experiments the crystals were outgassed in the following way. After a temperature rise with a rate $10\ \mathrm{K \cdot min^{-1}}$ in situ in a helium flow of $200\ \mathrm{ml \cdot min^{-1}}$, the sample was heated at 498 K for 2 hours in order to remove adsorbed impurities.

Helium was obtained as an ultra-high purity gas ($>$99.999%). The gaseous adsorbates such as methane, ethane, propane, *n*-butane, and i-butane were 3.5 grade ($>$99.95%).

The isotherm data were accurately measured in the temperature range from 303 to 408 K for methane,

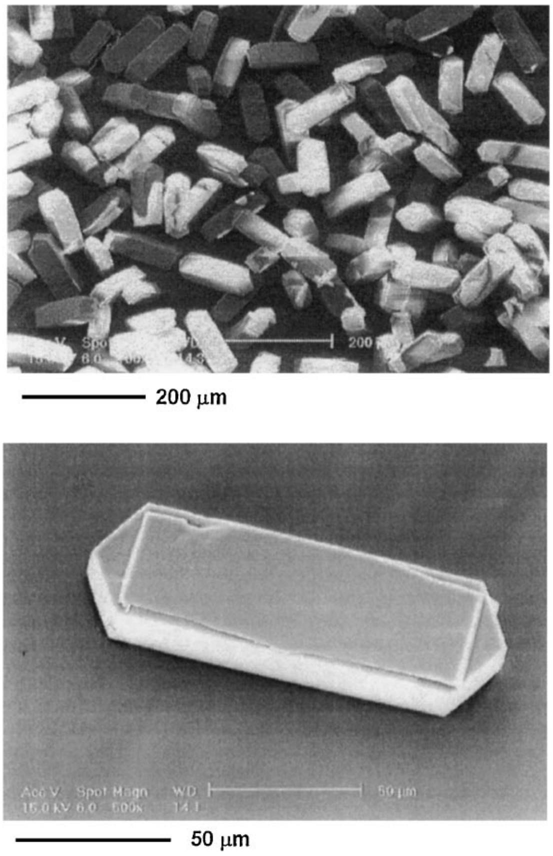

Figure 1. Scanning electron micrographs of the silicalite-1 sample used in this study.

ethane, and propane and from 303 to 473 K for the butane isomers. Six temperature levels were used for each *n*-butane or i-butane to enhance the accuracy of thermodynamic properties derived. Most experiments were repeated, and both adsorption and desorption experiments were performed to confirm reversibility.

Results and Discussion

Isotherms

The isotherms of the first four *n*-alkanes and i-butane adsorbed in silicalite-1 are shown in Figs. 2–6. The isotherms were reversible over the complete pressure range investigated. For the sake of clarity, the desorption points are omitted. The isotherms of the four *n*-alkanes exhibit a type-1 adsorption isotherm (Brunauer classification) over the temperature and pressure range studied. For the adsorption isotherm of i-butane at low temperatures a deviation from the type-1 behaviour is seen in Fig. 6. A 'kink' is observed for the isotherm obtained at 303 and 338 K. This has recently also been reported by Sun et al. (1998). In Fig. 7 results are given for isobars of *n*-butane and i-butane at 5×10^4 Pa over the temperature range of 303 to 525 K. A clear difference in the temperature dependence of the two isomers is observed.

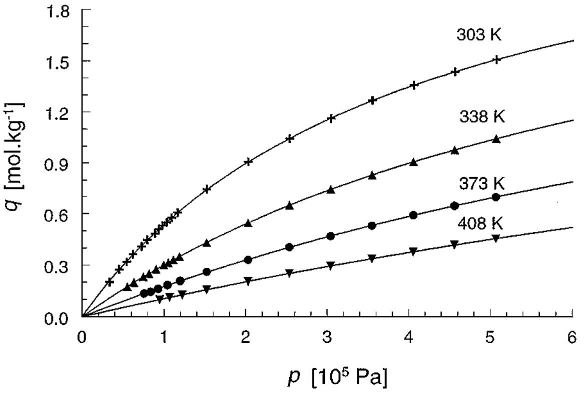

Figure 2. Adsorption isotherms of methane in silicalite-1. Lines are the Langmuir isotherm model fits.

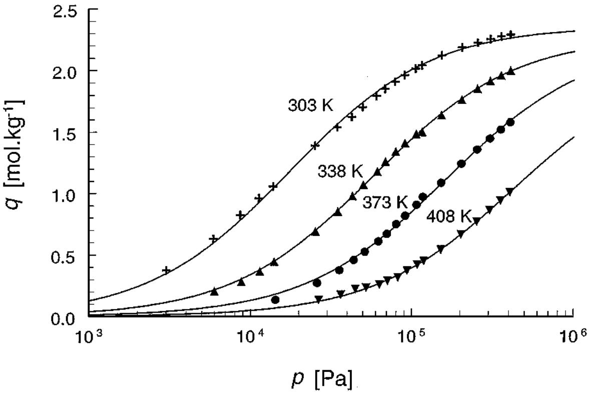

Figure 3. Adsorption isotherms of ethane in silicalite-1. Lines are the Langmuir isotherm model fits.

A comparison of our isotherm data with the literature data determined by different methods is compiled into Table 1. The measurement conditions of pressure and temperature and the measurement techniques are also given. Considering the differences in pressure and temperature, loadings of all the adsorbates investigated agree quite well with each other. Figure 8 also shows a comparison of our data and the data from Sun et al. (1998) for the butane isomers in the complete pressure range at 303 K. As is apparent in Fig. 8, our

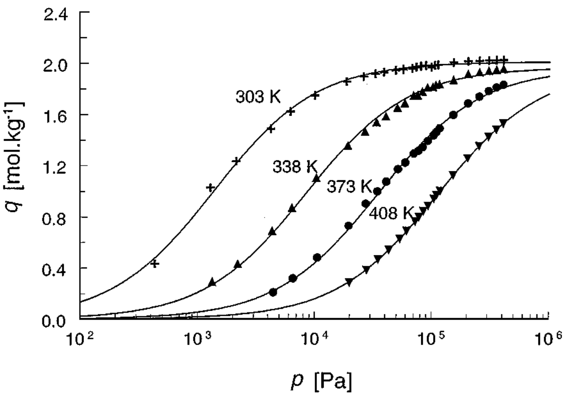

Figure 4. Adsorption isotherms of propane in silicalite-1. Lines are the Langmuir isotherm model fits.

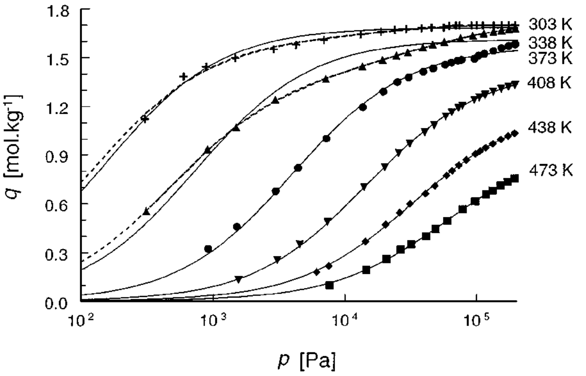

Figure 5. Adsorption isotherms of *n*-butane in silicalite-1. Lines are the Langmuir isotherm (solid) and the dual-site Langmuir isotherm (dashed) model fits.

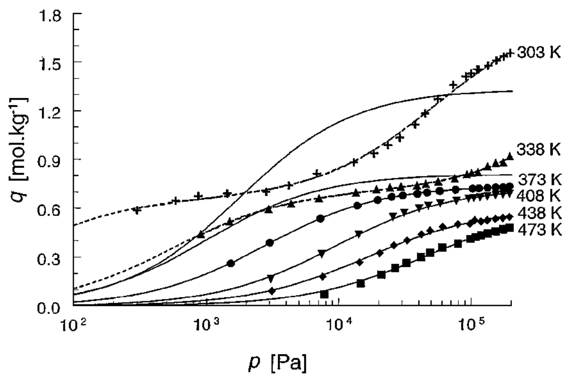

Figure 6. Adsorption isotherms of i-butane in silicalite-1. Lines are the Langmuir isotherm (solid) and the dual-site Langmuir isotherm (dashed) model fits.

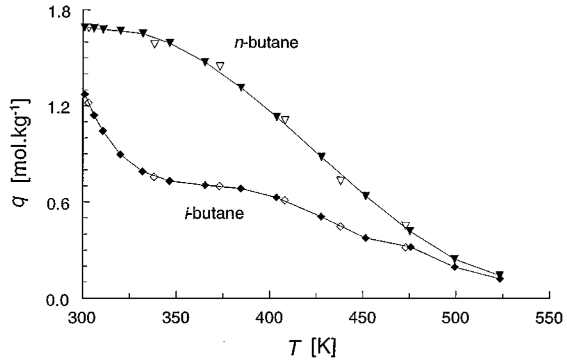

Figure 7. Isobars of *n*-butane and i-butane in silicalite-1 at a pressure of 5×10^4 Pa. Also given are isotherm results. Closed symbols: data from isobars; open symbols: data from isotherms.

results are in good agreement with the data from Sun et al. (1998), determined by the gravimetric method.

The adsorption equilibrium data of ethane and propane by Hampson and Rees (1993) with the isosteric method and of methane by Golden and Sircar (1994) with the volumetric method are also given in Table 1. Under the similar conditions, our results are also in good agreement with these literature data. Abdul-Rehman et al. (1990) measured the equilibrium adsorption of C_1-C_4 normal alkane on Linde S-115 silicalite with the volumetric technique. For methane and ethane, good agreement is seen in Table 1. However, for propane and *n*-butane, our results are much higher than the data from Abdul-Rehman et al. (1990). As pointed out by Sun et al. (1998), these differences may be related to the samples; pure crystals of uniform shape and size were used in this study while Abdul-Rehman et al. (1990) used silicalite-1 pellets with a binder. In addition, we found that the adsorption capacity of silicalite-1 is also related to the sample preparation. The remaining template in the sample can significantly decrease the adsorption capacity. Compared to our previous results of the same systems in silicalite-1 calcined at 723 K (Zhu et al., 1998), the capacities of the present silicalite-1 for the adsorbates investigated are higher by about 20%.

In general, there is a tendency for adsorbate molecules to locate in the smaller channels to maximise their attractive interactions with the silicalite-1. If the diameters of the adsorbed molecules are too large, this will lead to repulsion constraints and the molecules will reside in the larger intersections (June et al., 1990).

For the regular three-dimensional silicalite-1 pore structure it is instructive to express the preferential location in terms of molecules per unit cell. In silicalite-1 crystals each unit cell consists of four segments of

Table 1. Isotherm data: Comparison of different experimental techniques.

Method	T (K)	p (10^5 Pa)	q ($\mathrm{mol \cdot kg^{-1}}$) This study[a]	q ($\mathrm{mol \cdot kg^{-1}}$) Ref. data	Ref.
Methane					
Gravimetric	308	5.189	1.523	1.490	Sun et al., 1998
Volumetric	304	4.135	1.370	1.366	Golden and Sircar, 1994
Volumetric	300	5.545	1.567	1.509	Abdul-Rehman et al., 1990
Ethane					
Gravimetric	308	5.141	2.279	2.248	Sun et al., 1998
Isosteric	298	0.25	1.389	1.45	Hampson and Rees, 1993
Volumetric	300	2.900	2.223	1.838	Abdul-Rehman et al., 1990
Volumetric	298	0.87	1.936	1.55	Hufton and Danner, 1993
Propane					
Gravimetric	308	3.515	2.020	2.015	Sun et al., 1998
Isosteric	298	0.47	1.952	1.90	Hampson and Rees, 1993
Volumetric	300	3.424	2.002	1.590	Abdul-Rehman et al., 1990
n-Butane					
Gravimetric	308	1.034	1.699	1.632	Sun et al., 1998
Volumetric	300	1.150	1.700	1.386	Abdul-Rehman et al., 1990
i-Butane					
Gravimetric	308	1.026	1.420	1.371	Sun et al., 1998

[a] TEOM data at the same pressure as that in the literature and at 303 K.

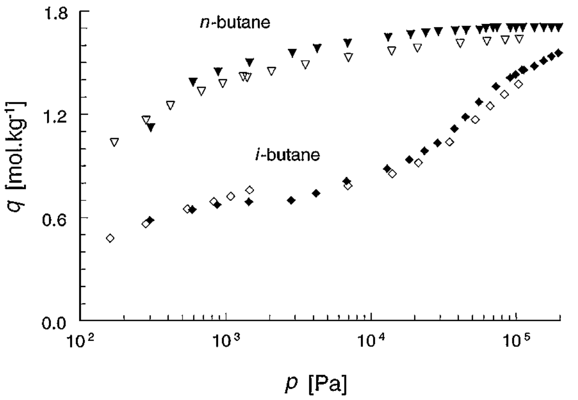

Figure 8. Comparison of *n*-butane and i-butane isotherm data to data from the literature. Closed symbols: this study at 303 K; open symbols: data at 308 K from Sun et al. (1998).

straight channels, four segments of zig-zag channels, and four segments of channel intersections (Richards and Rees, 1987). The size of an i-butane molecule, with a kinetic diameter of 0.5 nm, is comparable to the diameters of both the straight and the zig-zag channels. In this case the repulsion between i-butane molecules and the channel wall dominates over the attractive interaction between the adsorbate and the adsorbent. Therefore, i-butane molecules prefer to reside in the larger intersections.

Molecular simulations show that single-branched alkanes prefer to reside in the channel intersections and that the linear alkanes indeed reside in the channels (June et al., 1990; Smit et al., 1997). The difference between the channels and the intersections is much larger than the difference between the two types of channels. This means that one can say that essentially two different adsorption locations exist in silicalite-1.

To describe this difference of adsorbate location a dual-site Langmuir model has been proposed (Krishna et al. 1999; Micke et al., 1994; Millot et al., 1999; Vlugt et al. 1999; Zhu et al., 1998). This model takes into account the geometrical constraints of the zeolite pore structure and the different interaction potential in the channels and the intersections of the silicalite-1 crystals

$$q = q_{\mathrm{sat,C}} \frac{K_{\mathrm{C}} p}{1 + K_{\mathrm{C}} p} + q_{\mathrm{sat,I}} \frac{K_{\mathrm{I}} p}{1 + K_{\mathrm{I}} p} \qquad (2)$$

with the subscripts C and I refer to the channels and intersections, respectively. The geometrical constraints give rise to two different saturation amounts adsorbed in locations C and I and indicated by $q_{\mathrm{sat,C}}$ and $q_{\mathrm{sat,I}}$. In the case of low loadings, adsorbate molecules reside in the energetically preferred locations. In that case, the dual-site Langmuir model simplifies to the well-known

Langmuir model.

$$q = q_{\text{sat}} \frac{Kp}{1 + Kp} \tag{3}$$

The isotherms of methane, ethane, propane in silicalite-1 are well described by the conventional Langmuir model, as shown in Figs. 2–4. For the isotherms of n-butane in silicalite-1 only small deviations from the Langmuir model are found at 303 and 338 K (Fig. 5). With increasing loading, n-butane molecules gradually occupy the intersections, resulting in the deviation from the Langmuir model. Consequently, at the low temperatures the isotherms of n-butane are well described by the dual-site Langmuir model. The estimated saturation amounts of the normal alkanes in silicalite-1 for the Langmuir model are presented in Fig. 9. In the low temperature range, the fitted q_{sat} values stay reasonably constant for all the normal alkanes. However, at high temperatures q_{sat} is a function of temperature and slightly decreases with an increase in the operating temperature. This behaviour emphasises a difficulty in applying site concepts in a strict way. But the Langmuir model is generally used to describe adsorption isotherms. Barrer (1978) pointed out that the Langmuir model is often satisfactory for zeolites, due to compensation effects, even for more heterogeneous systems.

It is interesting to note that the isotherm for i-butane (Fig. 6) exhibits a 'kink' at 303 and 338 K. A preferential packing of 4 i-butane molecules per unit cell corresponds to a loading of 0.693 mol · kg^{-1} for the intersections. From the model fitting we have an estimated adsorption capacity for the intersections, $q_{\text{sat,I}}$, in good agreement with this value (Fig. 10). This explains the change in the adsorption behaviour of i-butane. For a loading above the saturation amount in the intersections the adsorbing molecules are forced at higher pressures into the straight or zig-zag channels. If the dual-site Langmuir model is fitted to the results obtained at a high temperature one of the two terms becomes negligibly small.

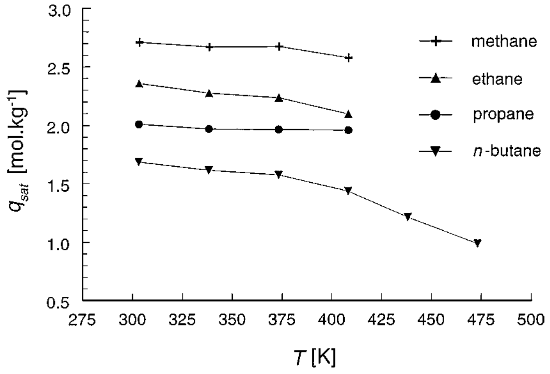

Figure 9. The saturation amounts adsorbed, fitted with the Langmuir model, for the normal alkanes as a function of temperature.

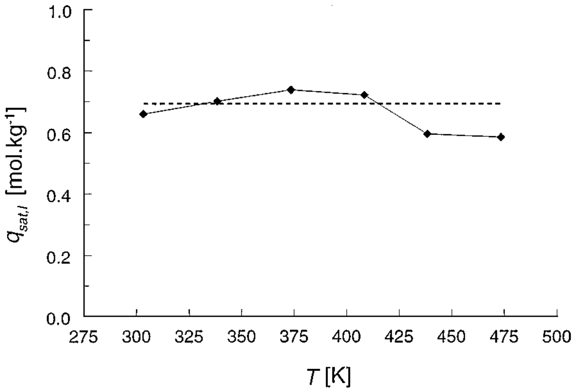

Figure 10. The i-butane adsorption capacity in the intersections, fitted with the dual-site Langmuir model for the isotherms at 303 and 338 K and the Langmuir model for the isotherms at 373, 408, 438 and 473 K, as a function of temperature. The dashed line represents a theoretical value of 0.693 mol · kg^{-1}.

Isobaric adsorption data of n-butane and i-butane in silicalite-1 are shown in Fig. 7. The n-butane isobar shows the typical trend obtained with the Langmuir isotherm model. The isobar for i-butane at 5×10^4 Pa exhibits a two-step behaviour. Over the temperature range of 303 to about 400 K, the isobar for i-butane is clearly different from the result obtained for n-butane. For the isobaric adsorption of i-butane, the decrease in the amount adsorbed is initially steeper with increasing temperature. This indicates that, if the temperature is increased, first the i-butane molecules residing in the channels desorb and that at higher temperatures the i-butane molecules are removed from the intersections. These results confirm the existence of the change of the adsorbate location with a change in the loading of i-butane in silicalite-1. Apparently, the large difference between the adsorption interaction for i-butane in the channels and in the intersections results in a clear two-step adsorption behaviour. This molecular location of i-butane in the micropores of silicalite-1 crystals was further confirmed by the simulation technique (Smit et al., 1997; Vlugt et al., 1998, 1999). A comparison with the isothermal results at the same pressure, obtained from different experimental runs, shows good agreement.

The various results, presented in Figs. 2 to 7, show that the temperature and the branching have a pronounced effect on the adsorption of light alkanes in silicalite-1. For the adsorption of *n*-butane and i-butane, the smallest alkane isomers, in silicalite-1 a molecular shape effect is observed.

Thermodynamic Properties

The isotherms have been used for the estimation of thermodynamic adsorption parameters. The isosteric heat of adsorption Q^{st} is defined by Eq. (4) (Ross and Olivier, 1964),

$$\left[\frac{\partial \ln p}{\partial T}\right]_q = \frac{Q^{\mathrm{st}}}{RT^2} \tag{4}$$

Q^{st} can be obtained from a plot of ln p vs $1/T$. Extrapolation then gives the isosteric heat of adsorption for the limiting case of adsorption at zero coverage. The calculated isosteric heat of adsorption at zero coverage is independent of the temperature. A comparison of the derived quantity Q_0^{st}, the isosteric heat of adsorption at zero coverage, is regarded as a useful way to check the temperature dependence of the initial slopes of the isotherms. The results in Table 2 show that the present data of the isosteric heat of adsorption agree well with chromatographic data (Hufton and Danner, 1993) and calorimetric results (Thamm, 1987).

Figure 11 shows the results of the isosteric heat of adsorption as a function of the amount adsorbed for the adsorbates investigated. For the normal alkanes, the values of Q^{st} almost stay constant in the complete range of the amount adsorbed. This indicates that silicalite-1 is energetically homogeneous for the adsorption of the normal alkanes. Figure 11 also gives the result of the isosteric heat of adsorption for i-butane at loadings up to 0.693 mol · kg^{-1}, corresponding to adsorption in the intersections.

Table 2. Isosteric heat of adsorption at zero coverage: comparison of experimental and literature data.

	$Q_0^{\mathrm{st}}[\mathrm{kJ \cdot mol^{-1}}]$		
Adsorbate	This study	G.C.[a]	Calor.[b]
Methane	17.7	20.4	
Ethane	32.4	29.8	32.0
Propane	43.6	40.5	
n-Butane	50.3	50.2	51.8
i-Butane	47.3	49.3	49.3

[a] Gas chromatographic method (Hufton and Danner, 1993).

[b] Calorimetric method (Thamm, 1987).

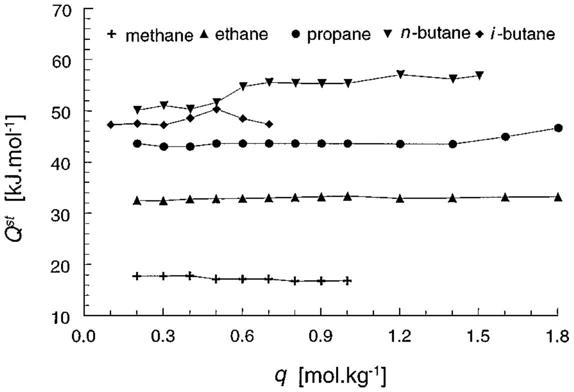

Figure 11. Isosteric heat of adsorption as a function of the amount adsorbed for all the adsorbates investigated in silicalite-1.

If Q^{st} and the adsorption enthalpy are assumed to be identical by neglecting any small temperature dependence, the differential molar entropy, $\bar{S}_{\mathrm{ads}}$, of adsorbates in the adsorbed phase can be calculated by Eq. (4) (Barrer, 1978),

$$\bar{S}_{\mathrm{ads}} = S_{\mathrm{g}}^0(298.15) + \int_{298.15}^{T} C_{\mathrm{p}} \frac{\mathrm{d}T}{T} + R \ln\left(\frac{p_0}{p}\right) - \frac{Q^{\mathrm{st}}}{T} \tag{5}$$

Here $S_{\mathrm{g}}^0(298.15)$ is the standard molar entropy of the gas phase at $T = 298.15$ K and $p_0 = 1.013 \times 10^5$ Pa. C_{p} is the molar heat capacity of gaseous adsorbates at constant pressure and T and p are the pressure and temperature under the equilibrium conditions. S_{g}^0 and C_{p} are obtained from the literature (Weast, 1987). Values of $\bar{S}_{\mathrm{ads}}$ were determined at each of the experimental temperatures for all the adsorbates and were plotted as a function of the amount adsorbed. The results at 303 K are illustrated in Fig. 12. The values of $\bar{S}_{\mathrm{ads}}$ increase with temperature and decrease with increasing loading. The curves of $\bar{S}_{\mathrm{ads}}$ at different temperatures of the individual components never intersect and follow parallel courses. For i-butane, a sharp decrease in the differential molar entropy in the adsorbed phase is seen as loadings up to the adsorption capacity in the intersections (0.693 mol^{-1} · kg).

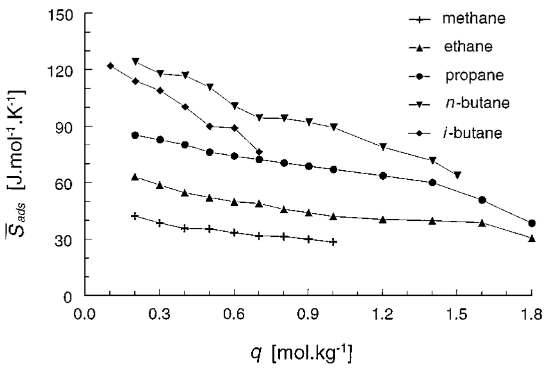

Figure 12. Differential entropy in the adsorbed phase as a function of the amount adsorbed for all the adsorbates investigated in silicalite-1 at 303 K. Points correspond to the data calculated from Eq. (5).

It is clear that entropy effects play a role in the preferential sitting in the intersections for i-butane. With an increase in the amount adsorbed above 0.693 $\text{mol}\cdot\text{kg}^{-1}$, the adsorbing molecules are forced into the channels. Therefore, more degrees of freedom are lost, resulting in an additional loss of the entropy. The same observation has been recently reported by Millot et al. (1999).

Conclusions

With the inertial microbalance technique, TEOM, it is possible to accurately measure equilibrium adsorption in microporous materials such as zeolites over a wide pressure and temperature range relevant to practical applications. The adsorption isotherms of methane, ethane, propane, *n*-butane, and i-butane in silicalite-1 have been obtained using this technique. Agreement with the literature data determined by other techniques indicates that the TEOM is a reliable technique. The isosteric heats of adsorption for all the adsorbates at zero coverage derived from the isotherm data are in good agreement with those determined by calorimetric and chromatographic methods.

The molecular geometry affects the adsorbate location within the micropores. By comparing the amount adsorbed with a preferential packing for i-butane, deviations from the Langmuir model have been explained. The isotherms of i-butane deviating from the Langmuir model can be well fitted by a dual-site Langmuir model, based on two adsorption locations. Adsorption of i-butane in silicalite-1 exhibits a two-step behaviour according to both isothermal and isobaric results. The adsorption of i-butane in silicalite-1 reveals the discrete-dual-structural heterogeneity for light alkanes in silicalite-1. Due to geometrical constraints i-butane first fills the intersections followed by filling of the channels. The molecular simulations support this picture.

Nomenclature

C_p	molar heat capacity ($\text{kJ}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$)
f_0, f_1	oscillation frequency (Hz)
K	equilibrium constant in the Langmuir isotherm model (Pa^{-1})
K_0	spring constant of oscillating element ($\text{kg}\cdot\text{Hz}^2$)
K_C, K_I	equilibrium constant in the dual-site Langmuir isotherm model (Pa^{-1})
p	pressure (Pa)
p_0	pressure of 1.013×10^5 Pa
q	amount adsorbed ($\text{mol}\cdot\text{kg}^{-1}$)
q_{sat}	saturation amount adsorbed in the Langmuir isotherm model ($\text{mol}\cdot\text{kg}^{-1}$)
$q_{sat,C}$, $q_{sat,I}$	saturation amounts adsorbed in the dual-site Langmuir isotherm model for the Channels and Intersections, respectively ($\text{mol}\cdot\text{kg}^{-1}$)
Q^{st}	isosteric heat of adsorption ($\text{kJ}\cdot\text{mol}^{-1}$)
Q_0^{st}	isosteric heat of adsorption ($\text{kJ}\cdot\text{mol}^{-1}$) at zero coverage
R	universal gas constant ($\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$)
$\bar{S}_{ads}$	differential molar entropy in the adsorbed phase ($\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$)
$S_g^0(298.15)$	standard molar entropy in the gaseous phase at 298.15 K and 1.013×10^5 Pa
T	temperature (K)

Greek Symbols

ΔM	total mass change (kg)
ΔM_a	adsorbed mass (kg)
ΔM_g	mass change caused by the change of the gas density (kg)

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