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A new Gaussian adsorption isotherm model: test data is methane-5A zeolite

Dana Abouelnasr · Kevin F. Loughlin

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Abstract An adsorption isotherm model is deduced based on the Gaussian cumulative frequency distribution principles. This isotherm has three parameters: q_{max}, the 50% adsorbate loading pressure P_{50}, and the standard deviation σ. The isotherm development hinges on establishing P_{50} and σ. These are deduced from a log plot of the isotherm pressure P versus the inverse Gaussian cumulative frequency distribution plot. The slope of this plot is σ and the y-intercept is P_{50}. The temperature dependency of P_{50} is deduced from a plot of log P_{50} versus $1/T$: this is actually the isosteric plot at 50% loading with slope equal to the heat of adsorption at 50% loading.

The model is tested using the literature methane data for 5A zeolite. Twenty-four isotherms from six different studies are fitted with the model using a q_{max} of 9 g/100 gZ applicable to methane 5A isotherms in the temperature range of the study, as reported in the literature. The standard deviation σ is observed to be 0.98 ± 0.088. The isostere of P_{50} indicates a heat of adsorption between 4.62–4.97 kcals/mole consistent with literature values. All 24 isotherms reduce to one characteristic curve when θ is plotted against $(P/P_{50})^{1/\sigma}$. The θ values approaching zero (<0.05) are shown to be consistent with reported Henry constant data using the heat of adsorption value calculated at 50% loading as is expected for homogeneous adsorption.

Keywords Isotherm · Gaussian model · Log normal probability distribution · Methane · 5A zeolite

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D. Abouelnasr · K.F. Loughlin (✉)
Department of Chemical Engineering, American University of Sharjah, PO Box 26666, University City, Sharjah, United Arab Emirates
e-mail: kloughlin@aus.edu

D. Abouelnasr
e-mail: dabouelnasr@aus.edu

Nomenclature

a	Constant in (10) and (16)
k	Boltzmann's constant
K	Langmuir equilibrium parameter, kPa^{-1}
K_{H}	Henry's constant, g/100 gZ/kPa
P	Pressure, kPa
P_{C}	Critical pressure, kPa
P_{LM}	Log mean pressure for the entire isotherm, kPa
P_{50}	Pressure at 50% loading, same value as P_{LM}, kPa
P_{r}	Reduced pressure, dimensionless
$P_{\mathrm{r},50}$	Reduced pressure at 50% loading, dimensionless
R	Gas constant
q	Zeolite loading, g/100 gZ
q_{max}	Maximum zeolite loading, g/100 gZ
T	Temperature, K
T_{CAR}	Critical adsorbate reduced temperature
T_{r}	Reduced temperature
Z	Standard normal probability distribution variable
u	Multisite Langmuir parameter

Greek Letters

θ	Loading ($=q/q_{\mathrm{max}}$)
σ	Standard deviation for the isotherm
$-\Delta H_0$	Heat of adsorption at 0% loading, kcals/mole
$-\Delta H_{50}$	Heat of adsorption at 50% loading, kcals/mole
Φ	Cumulative distribution function.

1 Introduction

Model isotherms have been used to characterize the adsorption of gases or liquids on adsorbents from late in the 19th century. Surveys of various isotherm models with their associated assumptions are published in many standard texts such as Do (1998) and Ruthven (1984). Standard isotherms are related to either surface coverage or micropore volume filling and are well covered in these textbooks.

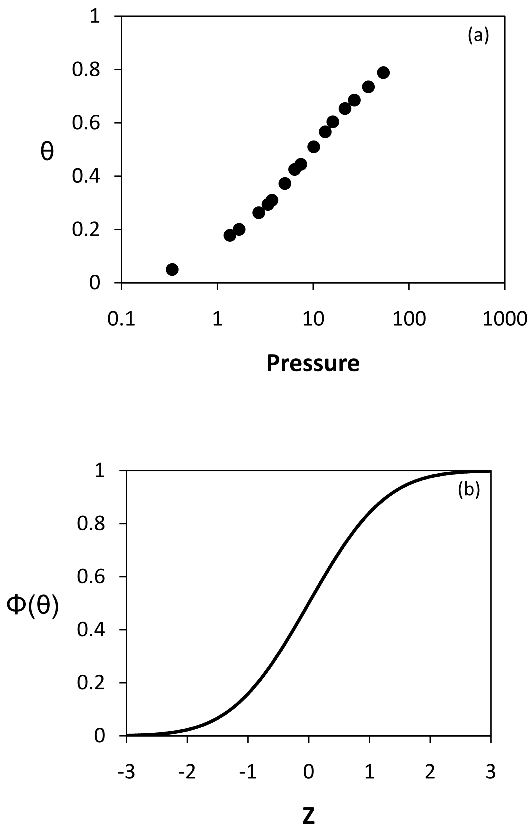


Fig. 1 (**a**) Isotherm for methane on 5A zeolite at 194.5 K. Data are from Stroud et al. A value for $q_{\max}$ of 9 g/100 gZ is used in calculating θ. (**b**) Cumulative distribution function for the standard normal distribution

The concept of normal probability distribution such as Gaussian has been much used in the adsorption literature. This usage has generally involved the use of typical isotherms with a superimposed Gaussian energy distribution. Examples are the employment of Gaussian energy distributions with Langmuirian (Braeuer et al. 1985, 1986; Jaroniec et al. 1987; Ruzic 1994 and Nouri and Haghseresht 2002), multilayer (Jaroniec et al. 1977; Panczyk and Rudzinski 2002), Fowler-Guggenheim (Morel et al. 1982), Jovanovic (Jaroniec and Piotrowska 1986), pore filling (Aharoni and Evans (1993), and various isotherms (Castro Luna et al. 1977; Garbacz et al. 1981; Derylo-Marczewska et al. 1987; Jaroniec and Choma 1988, and Cortes 1991). An alternative use for probability distributions is to describe the micropores in an activated carbon by either Gaussian or gamma distributions (John et al. 1985).

In this paper we propose a new isotherm in which the adsorption phenomena rather than the energy sites are modeled using a Gaussian normal probability distribution function.

2 Theory

The adsorption of gases onto adsorbents is a complex phenomenon. Gas-phase molecules must first contact an available site by colliding with the surface of the adsorbent. The molecule may then adsorb onto the site. Gas phase molecules possess a Maxwell-Boltzmann distribution of velocities and energies. If the molecules have sufficient kinetic energy, they may escape from the surface without adsorbing. So the probability that a molecule adsorbs onto the adsorbent surface depends upon the probability of collision with a site, the probability that the site is available, and the probability that the molecular kinetic energy is below a threshold to allow adsorption. A process which is dependent upon the probability of many different phenomena is often well-described by a log normal Gaussian distribution. Isotherms, when plotted as q or θ versus $\log P$ or $\log c$, exhibit the shape of the Gaussian cumulative frequency distribution. For example, a typical semi-log isotherm is shown in Fig. 1a for methane adsorption onto 5A zeolite. A cumulative normal distribution is shown beside it in Fig. 1b. The similarity in the two S-shaped curves suggests that the adsorption process does indeed follow the Gaussian cumulative distribution.

These observations allow the development of a new Gaussian isotherm with the following assumptions:

1. The molecules are assumed to have a Maxwell-Boltzmann distribution of velocities and energies in the adsorption system.
2. The adsorption micropore volume space of the adsorbent is uniform, that is, all the adsorption sites are equivalent, i.e., homogeneous.
3. The adsorbed species within the micropores is considered to behave as a liquid but with properties which may be different from those of a saturated liquid.
4. At the maximum adsorption, the adsorption space is filled to saturation.
5. There is no interaction with other adsorbed molecules which are located on the nearest neighbor adsorption sites.

The assumptions listed may need to be modified for a surface adsorbent. To relate the semi-log isotherm to the cumulative distribution, the standard normal probability distribution variable, Z, is first related to the log of the pressure, by (1):

$$Z = \frac{\log(P) - \log(P_{\mathrm{LM}})}{\sigma} \tag{1}$$

P_{LM} is the log-mean pressure for the entire isotherm. It is also the value of the pressure at 50% of the maximum loading, and so will be subsequently referred to as P_{50}. The standard deviation for the entire isotherm is specified as σ. Substituting for P_{LM} and rearranging (1) provides (2) and (3), which can be used interchangeably.

$$Z = \frac{\log(P) - \log(P_{50})}{\sigma} \tag{2}$$

$$Z = \log\left(\frac{P}{P_{50}}\right)^{\frac{1}{\sigma}} \tag{3}$$

The cumulative distribution function for the standard normal distribution, $\Phi(Z)$ is the area to the left of Z under the bell shaped curve. It corresponds to θ, the loading. For a standard normal distribution, the cumulative distribution function is defined by (4) (Walpole et al. 2002).

$$\Phi(Z) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{Z} e^{-z^2/2} dz \tag{4}$$

In standard adsorption notation, this equation is the Gaussian isotherm model equation:

$$\theta = \frac{q}{q_{\max}} = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\log(\frac{P}{P_{50}})^{1/\sigma}} e^{-z^2/2} dz \tag{5}$$

Instead of evaluating the integral, the value of the cumulative distribution function is more commonly determined from tables of the standard normal distribution. For simplicity, the cumulative distribution function is henceforth referred to as Φ, and the inverse function as Φ^{-1}. In Excel, functions NORMSDIST and NORMSINV can be used to calculate Φ and Φ^{-1}. Substituting for Z and setting $\Phi(Z)$ equal to θ provides (6):

$$\Phi\left[\frac{\log(P) - \log(P_{50})}{\sigma}\right] = \theta \tag{6}$$

Another form of this equation may be obtained by taking the inverse function of the cumulative distribution, shown in (7) and (8):

$$\left.\frac{\log(P) - \log(P_{50})}{\sigma}\right] = \Phi^{-1}(\theta) \tag{7}$$

$$\log\left(\frac{P}{P_{50}}\right)^{1/\sigma} = \Phi^{-1}(\theta) \tag{8}$$

Equation (7) can be rearranged as shown in (9).

$$\log(P) = \sigma\Phi^{-1}(\theta) + \log(P_{50}) \tag{9}$$

Equation (9) specifies a straight line for a plot of $\log(P)$ vs. $\Phi^{-1}(\theta)$, with a slope equal to the standard deviation, and a y-intercept equal to $\log(P_{50})$.

At a constant loading of 50% of the maximum, the pressure, P_{50} is related to the temperature by the isostere relation (De Boer 1968):

$$P_{50} = a\sqrt{T} e^{-\Delta H_{50}/RT} \tag{10}$$

where a is a constant and $(-\Delta H_{50})$ is the heat of adsorption at 50% loading. If the influence of the square root of the temperature is neglected, then this equation dictates that a plot of $\log P$ vs. $1/T$ should provide a straight line of slope $(-\Delta H_{50}/R)$. Although the influence of the square root of the temperature is small enough that a straight line can be perceived, this term will be maintained when data are fit to the isostere relation using regression techniques.

At low values of the adsorption loading, q is nearly proportional to the pressure, and Henry's Law applies:

$$K_{\mathrm{H}} = \lim_{p\to 0}\left(\frac{dq}{dp}\right) \tag{11}$$

We may substitute $(\theta q_{\max})$ for q, and $P_{50} * (P/P_{50})$ for P providing the adsorption is homogeneous giving (12):

$$K_{\mathrm{H}} = \frac{q_{\max}}{P_{50}} \lim_{p\to 0}\left(\frac{d\theta}{d(P/P_{50})}\right) \tag{12}$$

The above relationship, expressed in terms of the heat of adsorption, does not apply to adsorption on heterogeneous surfaces where the heat of adsorption at 50% loading is expected to be significantly different from that at 0% loading. Therefore, the Henry's Law constant for methane adsorption onto 5A zeolite, expected to be homogeneous, can be found from the slope of a plot of θ vs. P/P_{50}. Alternatively, if the standard deviation is not close to unity, then one may instead use the slope from a plot of θ vs. $(P/P_{50})^{1/\sigma}$ to determine the Henry's Law constant. The gradient may also be determined analytically by taking the derivative of (4) with respect to 10^Z, which provides (13).

$$\frac{d\theta}{d(P/P_{50})^{1/\sigma}} = \frac{d\Phi(Z)}{d(10^Z)} = \frac{e^{-Z^2/2}}{\sqrt{2\pi}\ln 10} \tag{13}$$

The gradient for this relation approaches zero as Z approaches $-\infty$, which is analogous to P approaching zero. This implies that $K_{\mathrm{H}} = 0$ in (11) which is similar to the Freundlich isotherm. In the results, the model [see (5)] is applied at very low loadings [up to $\theta \le 0.05$] to provide a nonzero value for K_{H}.

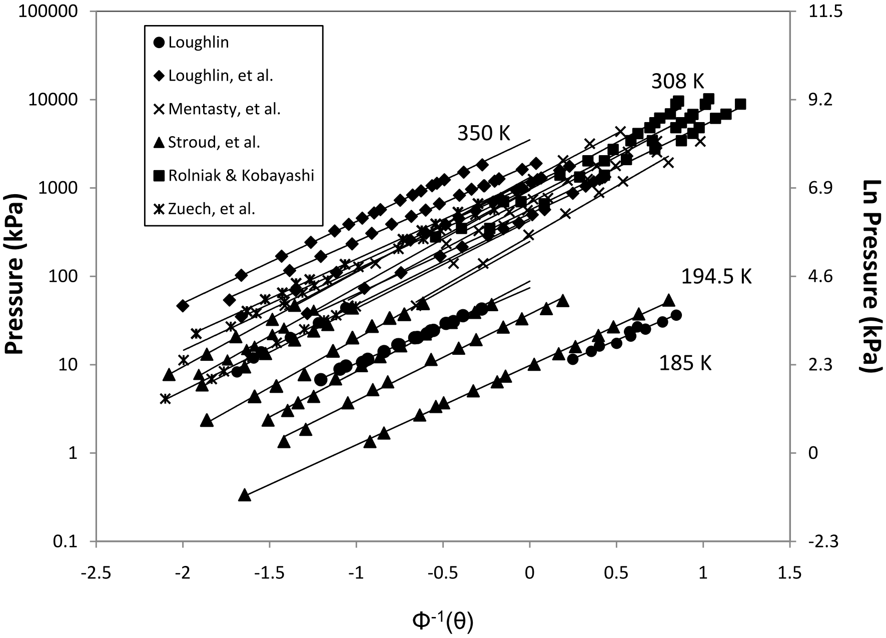
The isotherm has the interesting property in that it can be written in terms of reduced pressures with no change. Equations (1) to (9) above may be written in terms of pressure or reduced pressure producing the same result. This is because the introduction of P_{C} to form reduced pressures cancels as it is in both the numerator and denominator in all these equations. As an example, (14) below gives the exact same result as (6).

$$\theta = \frac{q}{q_{\max}} = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\log(\frac{P_r}{P_{r,50}})^{1/\sigma}} e^{-Z^2/2} dz \tag{14}$$

Equations (10) to (12) involve constants which need to be recalculated if reduced pressures are used.

One other observation that may be pertinent follows. In our calculations for methane on 5A zeolite, the standard deviation σ was found to be 1. This implies that only two parameters $q_{\max}$ and P_{50} are needed to specify the isotherms

Fig. 2 Linearized isotherms for methane adsorption on 5A zeolite. Data are from the studies listed in Table 1, using a $q_{\max}$ of 9 g/100 gZ



for this particular system. $q_{\max}$ may be determined independently (Loughlin and Abouelnasr 2009) leaving only one parameter P_{50} to be determined from the experimental data. This parameter has the interesting property that it is calculated at exactly $\theta = 0.5$, right in the middle of the isotherm. This is different from most present isotherms which are calculated using Henry's constants evaluated at zero loading, i.e., $\theta \sim 0.0$. It is not known at this time whether this difference will turn out to be significant.

3 Data collection

To validate the Gaussian model, it must be tested using experimental data. Data were collected from the literature for methane adsorption onto 5A zeolite. Six studies provided data for this work, and are listed in Table 1. The literature data used in this work include 24 isotherms at 17 different temperatures, covering a range from 185 K to 350 K, or a range of reduced temperatures from 0.97 to 1.84. As observed in Loughlin and Abouelnasr (2009), the critical adsorbed phase reduced temperature, T_{CAR}, for methane on 5A zeolite is 0.83. Adsorbed methane on 5A zeolite exhibits supercritical behavior above reduced temperatures of 0.83, so that all the available isotherms are for adsorbed phase supercritical methane. The saturation loading $q_{\max}$ for methane on 5A zeolite above a T_{CAR} of 0.83 was found to be (Loughlin and Abouelnasr 2009)

$$q_{\max} = 8 \pm 1 \frac{\mathrm{g}}{100\ \mathrm{g}} Z \tag{15}$$

Data from each study was converted to provide methane loadings in terms of grams of methane per 100 grams of zeolite crystal. For those studies that used pellets with a clay binder, this involved dividing the loading value by the fraction of crystal in the pellet. Each of the 24 isotherms covers only a portion of the entire S-shaped curve. Two of the more-complete isotherms are at 194.5 K and 308 K; since these two isotherms bracket most of the other isotherms, they will be used for specific comparisons between isotherm models.

4 Results and discussion

The experimental isotherm data was examined to determine whether it was consistent with the proposed Gaussian model. According to (9), a plot of $\log(P)$ vs. $\Phi^{-1}(\theta)$ should provide a straight line of slope σ and y-intercept of $\log(P_{50})$. θ was evaluated using a $q_{\max}$ of 9 g/100 gZ consistent with (15). The isotherms are plotted in linearized form in Fig. 2 as per (9). The resulting values of P_{50}, σ and correlation coefficients for all the isotherms are tabulated in Table 2. All isotherms fit the linear model well, with regression coefficients ranging from 0.9816 to greater than 0.999. The lines are practically parallel, with slopes ranging from 0.83 to 1.13, with a mean of 0.98 and standard deviation of 0.088. This indicates that the standard deviation is nearly constant for all the isotherms studied, and is assumed to be 1. If the linearized isotherm is plotted using natural logarithms, the slope will be larger by a factor of ln(10), giving a value of 2.3 for the slope. In contrast to the constant slopes, the y-intercept, P_{50}, increases dramatically with increasing temperature, due to the increasing pressures required to compensate for the effects of the exothermic nature of the adsorption process.

Table 1 Adsorption studies of methane on 5A zeolite

Author	Adsorption Temperature (K)
Loughlin (1970)	185, 230, 273
Loughlin et al. (1990)	275, 300, 325, 350
Mentasty et al. (1991)	258, 273.3, 283, 298
Rolniak and Kobayashi (1980)	288, 298, 308
Stroud et al. (1976)	194.5, 218, 233, 253, 273, 288, 300
Zeuch et al. (1983)	273, 298, 308

Table 2 Values for P_{50}, σ, and correlation coefficient for the 24 linearized isotherms of methane adsorption on 5A zeolite

Temperature (K)	P_{50} (kPa)	σ	R
185	7.2	0.83	0.9856
194.5	9.8	0.90	0.9995
218	37.4	0.98	0.9980
230	74.0	0.85	0.9988
233	88.2	1.02	0.9976
253	248	1.10	0.9966
258	283	1.13	0.9974
273	451	0.97	0.9991
	654	1.10	0.9959
	427	0.99	0.9980
273.3	514	0.96	0.9816
275	511	0.91	0.9984
283	673	1.05	0.9976
288	563	0.96	0.9981
	945	1.10	0.9988
298	773	1.00	0.9951
	990	0.92	0.9966
	1245	1.04	0.9988
300	1093	0.90	0.9997
	1313	1.07	0.9993
308	960	1.06	0.9944
	1226	0.89	0.9994
325	1825	0.86	0.9995
350	3501	0.92	0.9996
Average		0.98	0.9968
Std. Dev.		0.088	0.0044

The heat of adsorption can be estimated from the isostere relation for 50% loading, see (10). The 50% loading isostere is shown in Fig. 3. Neglecting the influence of the square root of the temperature, the heat of adsorption is estimated as the slope of the linear regression line through the data, and is 4.97 kcal/mole. When regression techniques are used with (10), such that the influence of the square root of the temperature is included, a slightly different value is observed for the heat of adsorption: 4.62 kcal/mole. These estimates of the heat of adsorption are very similar to the previously-reported values of 4.5–5.12 kcal/mole reported in the literature by Loughlin et al. (1990). The best-fit value

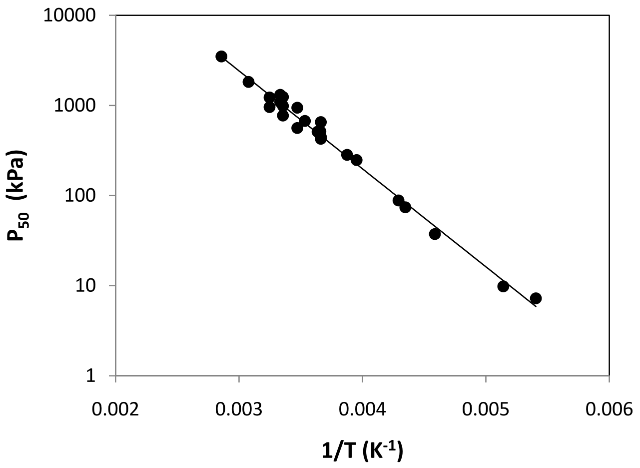


Fig. 3 Isostere for a constant 50% loading. P_{50} values are determined from experimental data from the 24 isotherms listed in Table 1

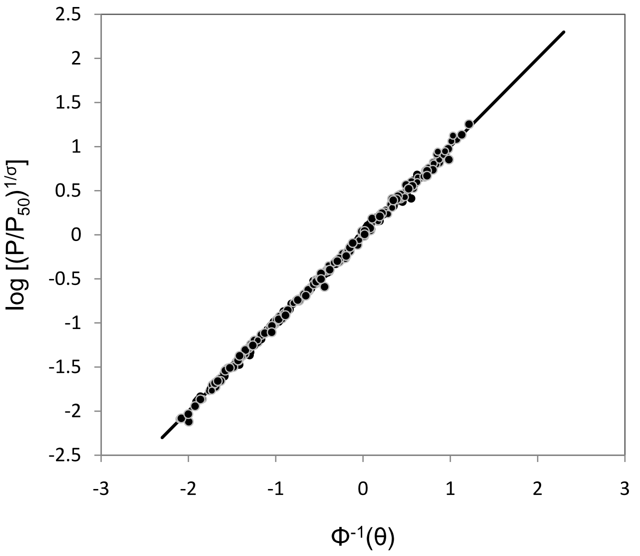


Fig. 4 Data from 24 methane isotherms, overlaid on a single normalized linear isotherm. Values for P_{50} and σ are parameters fitted to each isotherm separately. Data are from the studies listed in Table 1, using a $q_{\max}$ of 9 g/100 gZ

of 4.62 kcal/mole, along with a best-fit value for the pre-exponential factor a of 1.35×10^5 may be used to model the pressure at 50% loading as a function of temperature.

$$P_{50}(\text{kPa}) = 1.35 \times 10^5 \sqrt{T} e^{-\Delta H_{50}/RT} \tag{16}$$

All 24 isotherms can be overlaid onto a single line, by normalizing the lines, as described by (8). Accordingly, a plot of $\log(P/P_{50})^{1/\sigma}$ vs. $\Phi^{-1}(\theta)$ provides a single straight line of slope 1 which passes through the origin. All of the experimental data are well-described by this line, as is displayed in Fig. 4.

The data can also be shown in a more familiar form as one characteristic curve in Fig. 5, by plotting θ vs. the normalized pressure term, $(P/P_{50})^{1/\sigma}$, so that the S-shaped curve returns, as described by (6). The data from all 24 isotherms are displayed on a single curve, indicating that they are well-described by the model. Finally, the experimental data may be shown in its customary form, as the loading q vs. pressure. This is illustrated in Fig. 6, where each isotherm can be seen separately, and the model for each isotherm is drawn as a solid line. Once again, the model is very consistent with the experimental data for each isotherm. An interesting observation is that the empty V shaped space shown in Fig. 6 is an illustration of the different experimental techniques used to measure the data. The data to the left of the empty V shape is measured using gravimetric glass apparatus which restricts the maximum pressure to 100 kPa. The isotherms to the right of the empty V shape are measured using volumetric, chromatographic or column techniques which may be measured to a much higher pressure.

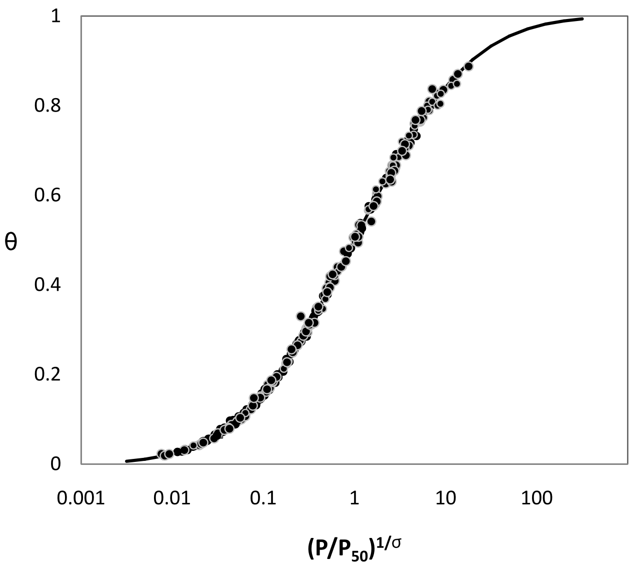


Fig. 5 Data from 24 methane isotherms, overlaid on a single normalized isotherm graph. Values for P_{50} and σ are parameters fitted to each isotherm separately. Data are from the studies listed in Table 1, using a $q_{\max}$ of 9 g/100 gZ

A value for the Henry's Law constant, K_H, can be estimated from the slope of the normalized isotherm below a θ of 0.05, so that several temperatures may contribute to the determination of the slope. Note that this can only be done for homogeneous adsorption for which methane on 5A zeolite is established. In Fig. 7, θ is plotted as a function of normalized pressure using data taken from Fig. 6 for θ below 0.05; note that this is not a semi-log plot. The best-fit line passing through the origin provides a slope of 2.28. Equation (13) provides an analytical determination of the gradient, which ranges from 2.5 to 2.0 for a range of theta from 0.01 to 0.05. Since the experimentally-observed value of 2.28 is approximately mid-range, this value will be used for further calculations. From (12), K_H is equal to the slope

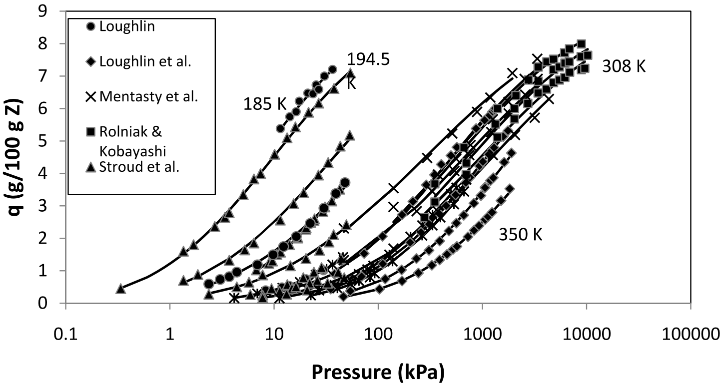


Fig. 6 Data from 24 adsorption isotherms for methane. The Gaussian model for each isotherm is shown as a *solid line*. Data are from the studies listed in Table 1

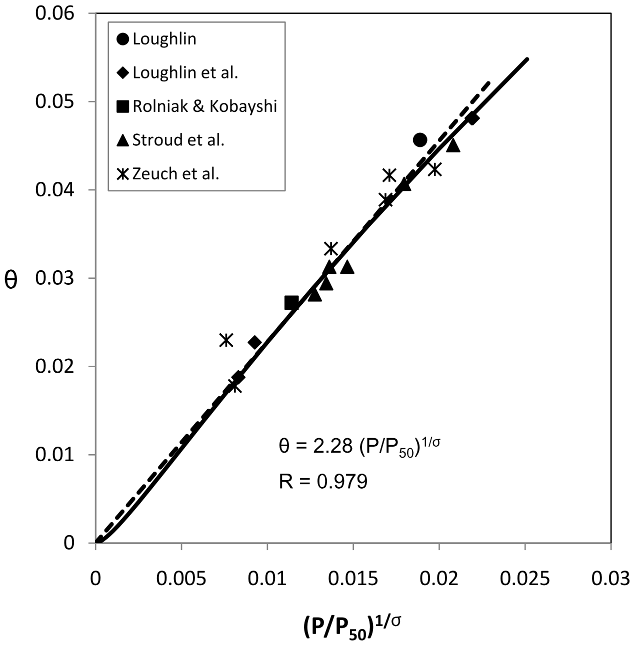


Fig. 7 Determination of slope for Henry's law constant determination. The *solid line* represents the Gaussian model. The *dotted line* is a linear regression of the data for theta less than 0.05

multiplied by $q_{\max}$, and divided by P_{50}, provided by (10). This gives the following relation for the dependence of K_{H} on temperature.

$$K_{\mathrm{H}}\left(\frac{\mathrm{g}}{100\ \mathrm{g\,kPa}}\right)=\frac{1.52\times10^{-4}}{\sqrt{T}}e^{-\Delta H_{50}/RT} \tag{17}$$

This relation is compared to previously-reported values for the Henry's Law constants for several temperatures reported by Loughlin et al. (1990) in Fig. 8. The figure indicates that the model for Henry's Law constant derived from the Gaussian model is consistent with the values previously observed. Also, the excellent fit verifies the homogeneity of the system as the heat of adsorption at 0 ($-\Delta H_0$) and 50% ($-\Delta H_{50}$) loading appears similar.

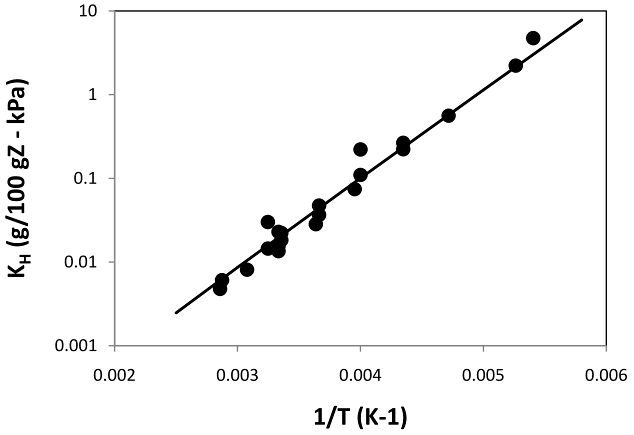


Fig. 8 Henry's Law relationship derived from the Gaussian model (*solid line*) to previously-reported values of Henry's Law constants. Values are taken from Table 5 of Loughlin et al. (1990), calculated from different studies

The performance of the Gaussian isotherm model is compared to that of other isotherm models in Figs. 9 and 10. Figure 9 shows the two most complete isotherms, at temperatures of 194.5 and 308 K. The other isotherms have been removed for clarity. The experimental data are shown with three models; the Gaussian model as a solid line, the Langmuir model as a dashed line, and the multi-site Langmuir (MSL) model as a dotted line. The constants used in calculating the Langmuir and multisite Langmuir isotherms were determined from regression methods, and are presented in Table 3. At 194.5 K, both Langmuir and MSL models underpredict q at the low-pressure end of the isotherm, and overpredict q at the high-pressure end, although the MSL model is better than the Langmuir. At 308 K, the MSL model is much closer to the experimental data than the Langmuir model, and is similar to the Gaussian model. To aid in comparison of the models, the residuals for all three models are plotted in Fig. 10a for 194.5 K and Fig. 10b for 308 K. At 194.5 K, the maximum residual for the Gaussian model is 0.1 g/100 gZ, compared to a maximum of 0.55 for the Langmuir model and 0.31 for the MSL model. At 308 K, the

Table 3 Parameters used and resulting correlation coefficients for fitting the Langmuir and Multi-site Langmuir models to the isotherm data

Temperature	Langmuir		Multi-site Langmuir		
(K)	K (kPa^{-1})	R	K (kPa^{-1})	u/k (K)	R
194.5	0.105	0.984	0.112	−2	0.989
308	0.00079	0.919	0.0033	−700	0.995

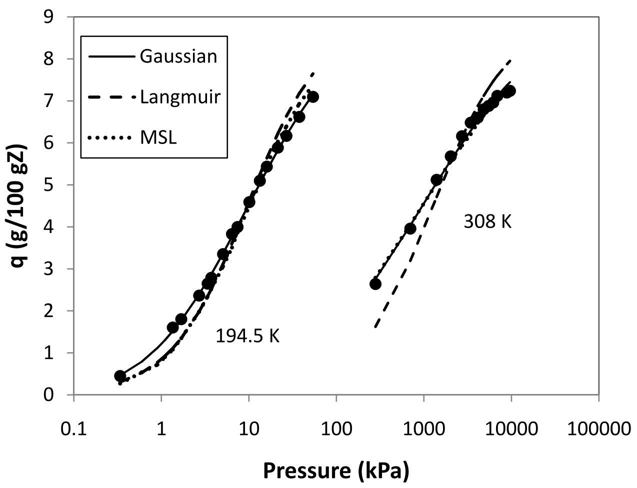


Fig. 9 Two isotherms of methane adsorption, 194.5 K from Stroud et al. (1976) and 308 K from Zeuch et al. (1983). The performance of the Gaussian isotherm model is compared to the Langmuir and the mult-site Langmuir isotherm models—see Table 3 for parameters

residuals for the Gaussian model and the MSL model are very similar, and both have similar values for the maximum residual, 0.20 g/100 gZ for the Gaussian model and 0.27 for the MSL model. The Langmuir model has much larger residuals, with a maximum of 1.0 g/100 gZ.

The adsorption behavior for methane in the temperature range of this study can be predicted using the Gaussian model, see (6), the mean value for the standard deviation, and (16) to estimate P_{50}. In this way, the experimental data for the 24 isotherms were compared to the predictions for each temperature. As shown in Fig. 11, the predictions fit the data reasonably well, although there is some spread of the data; some isotherms are slightly over-predicted, others slightly underpredicted, and still others fall on the Gaussian model curve, indicating good predictions.

5 Conclusions

The theory for a new Gaussian isotherm model is developed. The primary parameter of the new model is P_{50} which is the isotherm pressure at 50% saturation loading. The other primary parameter, the standard deviation is invariant and equal to 1 for the system studied, The new model isotherm is shown to be very consistent with experimental data for supercritical methane on 5A zeolite.

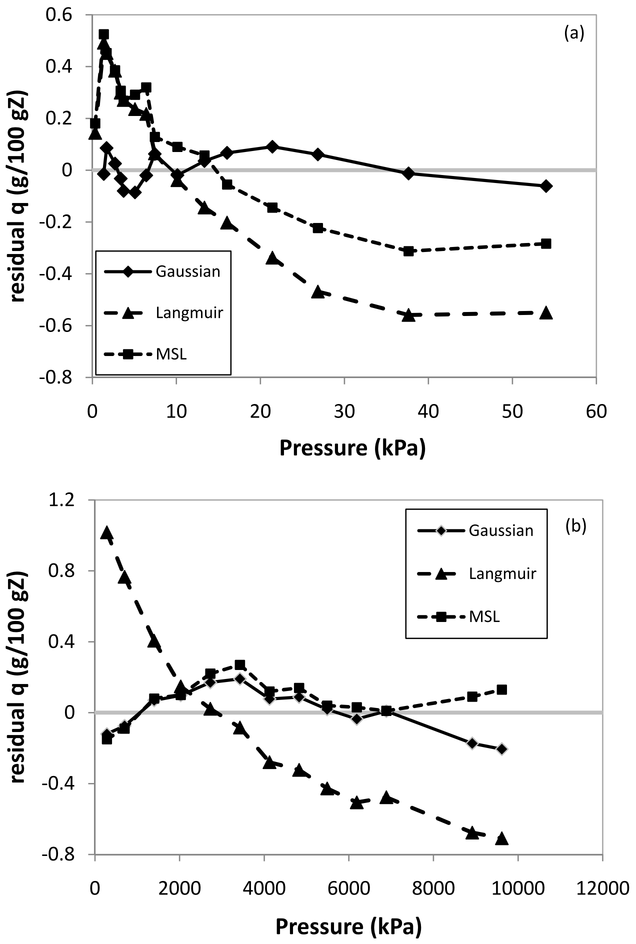


Fig. 10 At 194.5 K (Fig. 10a) and 308 K (Fig. 10b), the residuals for the three isotherm models: the Gaussian; Langmuir; and multi-site Langmuir (MSL)

An equation for P_{50} may be derived from the isostere at 50% loading. From this equation the heat of adsorption at 50% loading is calculated which is consistent with reported values. Also, as the system examined is homogeneous, the Henry constants are calculated from this heat of adsorption and from the P_{50} value, and are shown to be consistent with reported values in the literature.

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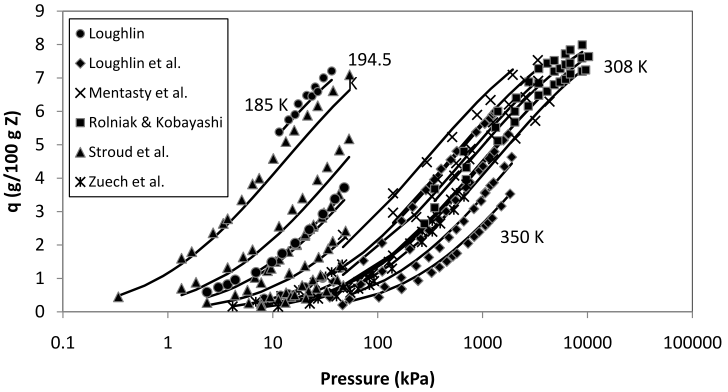
Fig. 11 Experimental data for 24 isotherms, compared to prediction from Gaussian model. Data are from studies listed in Table 1



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