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Adsorption Equilibrium of Carbon Dioxide and Methane on *β*-Zeolite at Pressures of Up to 2000 kPa Using a Static Volumetric Method

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The development of adsorption-based technologies for gas separation requires knowledge of adsorption equilibria on a specific adsorbent material. In this work, the adsorption equilibria for CO_2 and CH_4 on the β-zeolite adsorbent were determined at (308.1, 318.1, and 328.1) K over the pressure range of (0 and 2000) kPa using a static volumetric apparatus. Experimental data were correlated by the Langmuir, Sips, and Toth equations. Despite the relative simplicity of these models, the experimental data were fit very well. The preferential adsorption capacity of CO_2 on β-zeolite was much higher than that for CH_4, with the ideal selectivity of carbon dioxide over methane ranging from 2 to 7.

Introduction

β-Zeolite is one kind of high-silica microporous material with a pore of 0.71 nm × 0.73 nm, a typical Si/Al ratio of 10 to 25, and a three-dimensional 12 ring interconnected channel structure.[1] It is of great interest as a promising catalyst or separator for various separation and reaction processes because of its preferential adsorption of one component over another.[2−4] Thus, a number of adsorption investigations have been carried out on these type of zeolites for numerous compounds.[5−7] These studies have reflected that β-zeolite performs very well in separating C_5−C_8 alkane isomers or effectively discriminating the monobranched isomer from the dibranched one, because of its sinusoidal pore structure and selective adsorption capacity.[5,6] Furthermore, β-zeolite is also shown to be a good candidate for applications in flue gas separations, as well as natural gas and landfill gas purifications.[7] Hence, β-zeolite has recently received attention in preparing polymer-based composite membranes for gas separation applications.[8−14] Very promisingly, β-zeolite incorporated polyethersulfone (PES) composite membranes have exhibited pronounced permeation performance with an ideal selectivity of CO_2 over CH_4 of over six times greater than that of pure PES.[13] The progress of these composite membranes suggests that the performance enhancement gained may be due to the addition of Knudsen diffusion and selective adsorption to the solution-diffusion mechanism that predominates in nonporous pure polymer membranes.[14] These advances in separation and catalysis processes encourage equilibria and kinetic adsorption studies of various species of interest on β-zeolite for its subsequent potential applications.

Adsorption equilibria are the key information for the design of practical separation processes based on adsorption mechanisms. Up to now, there are few isotherm data for β-zeolite adsorbent at high pressure. These high-pressure equilibria data for single and multicomponent adsorption are needed to design a separation process such as a pressure-swing adsorption (PSA). However, multicomponent adsorption equilibria data are difficult to obtain directly and possibly predicted from single-component isotherm information.[15,16] Besides, the pure compound adsorption information may be useful to understand the enhanced separation performance of polymer composite membranes incorporated by β-zeolite entities.[8−14] Therefore, basic information about the adsorption equilibrium behavior of the pure components is required for a wide range of experimental conditions.

In this work, we have volumetrically measured the high-pressure adsorption equilibrium of methane and carbon dioxide on β-zeolite adsorbent. The adsorption experiments were carried out at (308.1, 318.1, and 328.1) K for methane and CO_2 and at pressures of up to 2000 kPa. The full set of experimental data was correlated by the Langmuir, Sips, and Toth equations.

Experimental Section

Materials. The adsorbates used were carbon dioxide and methane, and their purities were 99.99 % and 99.995 %, respectively. Both gases were purchased from Beijing Ya-Nan Gas Pte. Ltd. The adsorbent employed was β-zeolite, which was synthesized in our laboratory with a molar Si/Al ratio of 16:1.[11] Its properties were examined by using powder X-ray diffraction (XRD) analysis on a Shimadzu XRD-6000 spectrometer, Brunauer−Emmett−Teller (BET) measurements on Quantachrome AS-1 autosorb-1, and scanning electron microscopy (SEM) analysis on a JEOL JSM-6700F instrument.

Experimental Setup. Adsorption equilibrium of pure gases was performed by using a simple dual-volume adsorption apparatus. A schematic diagram of the experimental setup is shown in Figure 1. It mainly consists of an adsorption unit (AU), a dosing cell (DC), two pressure sensors (PS), a conventional oven, a network of stainless steel tubes and on−off valves, two filters (F), a vacuum pump (VP), a gas reservoir (GR), and a pressure regulator.

The adsorption apparatus was mainly located in a temperature controllable oven (± 0.1 K). Evacuation of the adsorption unit was done with a vacuum pump (VP) (Edwards RV5, U.K.) to realize a vacuum level less than 0.2 Pa. The regeneration of the sample inside the adsorption cell was carried out by venting off the gas, vacuuming for 4 h, and then purging helium gas for 1 h, followed by vacuuming for another 12 h. An on−off

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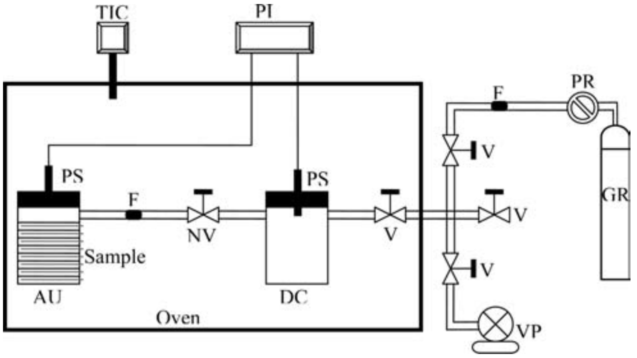

Figure 1. Experimental setup for zeolite adsorption measurements: PI, digital pressure indicator; TIC, digital temperature indicator and controller; AU, adsorption unit; DC, dosing cell; PS, pressure sensor; NV, needle valve; V: valve; F, 0.5 μm pore-size filter; VP, vacuum pump; PR, pressure regulator; GR, gas reservoir.

needle value was employed to connect the dosing cell and the adsorption cell (made in-house) to allow the introduction of the gas into the adsorption cell. A pressure sensor (Baratron 750B, MKS Instruments, USA) was applied to precisely measure the pressure from vacuum to 2000 kPa.

Experimental Procedure. The adsorbent sample was initially pressed in the form of discs, activated at 623.1 K in an electronic furnace overnight, and then dried at 473.1 K in a vacuum oven (< 1 Pa) for more than 12 h to remove any impurities retained inside. The regenerated sample of about (2 to 3) g was weighed using an analytical balance (Mettler AE200, ± 0.01 mg) and then loaded in the adsorption cell. After being housed in an oven for a constant testing temperature, the whole system (including the adsorption cell, the sample, and the dosing cell) was activated under vacuum for at least 24 h using the vacuum pump. An appropriate quantity of gas, supplied from a gas reservoir by using a pressure regulator, was fed into the dosing cell with a known volume. After reaching thermal and mechanical equilibria, an initial pressure was measured and recorded. By opening and then closing the needle valve, the gas was directed to the adsorption cell, and adsorption occurred. As in the first step, the pressures in the adsorption and dosing cells were measured after the achievement of adsorption equilibrium. An additional amount of the measured gas was continuously fed into the dosing cell from the gas reservoir to increase the pressure to a higher value for subsequent adsorption equilibrium measurements. The same set of operations was repeated to cover the whole pressure range at a constant temperature. It should be mentioned that the measurement intervals were long enough for at least 12 h, sometimes more than 24 h, to guarantee that the gas sorption in the samples fully reached an equilibrium state at the tested pressures.

The amount of the adsorbate, q, adsorbed on the adsorbent at a given temperature was calculated according to the mass balance equation that was derived from the generalized equation of state before and after adsorption equilibrium.[17]

$$\left.\frac{PV}{ZRT}\right|_{\mathrm{AU}}^{1} + \left.\frac{PV}{ZRT}\right|_{\mathrm{DC}}^{1} = \left.\frac{PV}{ZRT}\right|_{\mathrm{AU}}^{2} + \left.\frac{PV}{ZRT}\right|_{\mathrm{DC}}^{2} + \frac{q}{M} \tag{1}$$

where P and T are the experimental pressure and temperature, respectively, M is molecular weight of the adorbate gas, R is the universal gas constant, and Z is the compressibility factor obtained from the $P-V-T$ data.[18,19] V is the volume, and the volumes of the adsorption and loading cells were (108.61 and 122.73) cm^3, respectively, as determined from the expansion of helium gas. Superscripts 1 and 2 represent the state before and after adsorption equilibrium, respectively. Subscripts AU and DC stand for the adsorption unit and dosing cell, respectively.

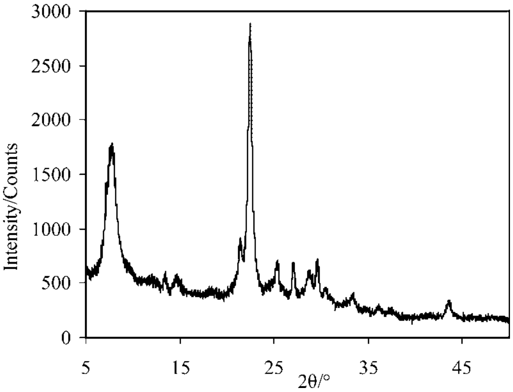

Figure 2. XRD spectrum of the adsorbent β-zeolite particles used.

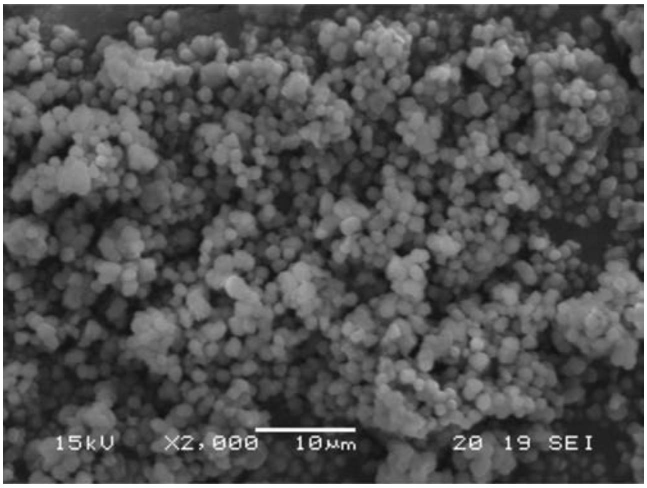

Figure 3. SEM image of the adsorbent β-zeolite particles used.

Results and Discussion

Adsorbent Properties. The XRD pattern of the adsorbent used here is shown in Figure 2, which is identical to those reported elsewhere.[11] As evidenced by the characteristic XRD peak spectra, the adsorbent particles have been confirmed to be a pure β-zeolite structure without other zeolite structures. The scanning electron microscopy (SEM) image of these particles is shown in Figure 3, and β-zeolite crystals exhibit an average particle size of 1.0 μm with a very narrow particle size distribution ranging from (0.5 to 1.5) μm. The micropore volume and total pore volume of the sample are (0.24 and 0.38) $\mathrm{cm}^3\cdot\mathrm{g}^{-1}$, respectively. The specific BET surface area is 608 $\mathrm{m}^2\cdot\mathrm{g}^{-1}$, where the external surface area is 117 $\mathrm{m}^2\cdot\mathrm{g}^{-1}$ and the micropore surface area is 491 $\mathrm{m}^2\cdot\mathrm{g}^{-1}$.

Gas Adsorption. In this study, equilibrium adsorption for carbon dioxide and methane onto β-zeolite was conducted at (308.1, 318.1, and 328.1) K and at pressures up to 2000 kPa. The experimental results are presented in Tables 1 and 2 and graphically shown in Figures 4, 5, and 6. Of the five types of physical adsorption according to the IUPAC classification, the experimental data illustrated in the above figures belong to the I-type Langmuir isotherm, characteristic of microporous adsorbents with small pore sizes. All of the isotherm curves showed nonlinearity, possibly attributed to the pore surface heterogeneity and active sites of the adsorbent, as well as adsorbate−adsorbent interactions. The adsorbent zeolite used is rich in aluminum content, which renders the pore surface hydrophilic. This is also

Table 1. Experimental Data of CO_2 Adsorption Equilibrium on *β*-Zeolite

$T = 308.15$ K		$T = 318.15$ K		$T = 328.15$ K	
P/kPa	q/mol·kg^{-1}	P/kPa	q/mol·kg^{-1}	P/kPa	q/mol·kg^{-1}
0.000	0.000	0.000	0.000	0.000	0.000
9.308	0.064	29.475	0.497	26.200	0.332
30.337	0.353	42.747	0.616	36.542	0.475
54.736	0.647	58.950	0.993	52.400	0.664
65.413	1.202	71.016	1.115	54.813	0.786
79.634	1.565	85.495	1.232	65.500	0.892
109.282	1.819	97.905	1.305	80.669	1.006
165.474	2.101	111.005	1.368	99.284	1.146
198.396	2.258	137.205	1.545	122.037	1.167
261.828	2.444	178.574	1.757	127.553	1.303
318.710	2.582	216.495	1.834	172.369	1.488
345.254	2.597	237.869	2.003	201.327	1.607
414.374	2.723	361.285	2.271	230.284	1.726
436.093	2.725	461.603	2.442	262.000	1.733
537.962	2.820	561.922	2.614	327.500	2.007
560.715	2.823	682.752	2.719	406.100	2.074
661.551	2.907	803.583	2.825	448.848	2.233
801.342	2.987	871.840	2.917	542.616	2.290
846.502	3.018	1036.280	2.962	675.685	2.507
944.752	3.098	1235.538	3.090	736.703	2.519
1043.003	3.177	1416.870	3.243	950.096	2.734
1161.678	3.262	1595.444	3.317	1097.643	2.853
1280.354	3.347	1675.216	3.334	1195.549	2.898
1340.338	3.345	1758.977	3.350	1416.525	2.984
1456.860	3.461	1829.336	3.367	1476.854	3.043
1542.699	3.509	1902.509	3.384	1671.286	3.075
1640.777	3.524			1754.850	3.106
1738.855	3.540			1842.593	3.137
1791.020	3.575			1934.723	3.168
1862.661	3.593				
1974.421	3.611				

Table 2. Experimental Data of CH_4 Adsorption Equilibrium on *β*-Zeolite

$T = 308.15$ K		$T = 318.15$ K		$T = 328.15$ K	
P/kPa	q/mol·kg^{-1}	P/kPa	q/mol·kg^{-1}	P/kPa	q/mol·kg^{-1}
0.000	0.000	0.000	0.000	0.000	0.000
24.821	0.080	27.407	0.098	61.363	0.093
56.192	0.196	85.495	0.223	88.597	0.174
72.395	0.230	151.512	0.385	159.958	0.277
106.351	0.342	208.911	0.529	252.692	0.446
165.129	0.471	289.407	0.647	380.590	0.633
188.227	0.535	347.495	0.720	486.424	0.740
271.308	0.703	443.332	0.826	588.122	0.849
328.879	0.786	511.590	0.921	722.397	0.949
363.698	0.848	601.739	1.009	849.260	1.044
421.269	0.933	699.989	1.097	962.162	1.111
497.973	1.035	800.308	1.178	1077.649	1.177
598.981	1.131	938.202	1.250	1168.142	1.218
668.273	1.190	1039.383	1.310	1258.119	1.257
820.819	1.304	1205.316	1.398	1363.608	1.338
993.533	1.429	1304.141	1.454	1495.125	1.390
1111.778	1.502	1401.012	1.493	1606.303	1.422
1273.459	1.580	1512.362	1.528	1695.073	1.452
1464.271	1.638	1699.899	1.587	1786.325	1.463
1659.220	1.743	1813.216	1.622	1845.258	1.475
1834.002	1.783	1942.054	1.631	1974.436	1.494
1930.303	1.801				

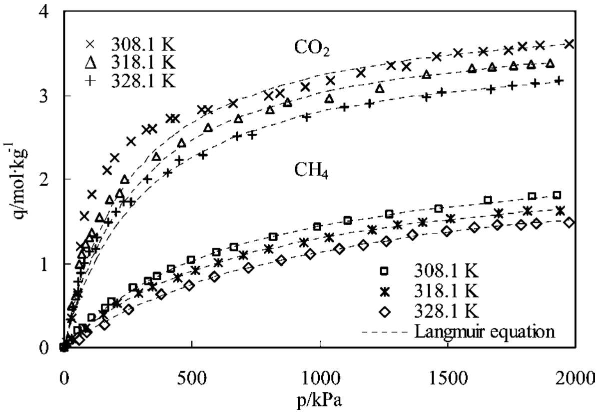

Figure 4. Predicted CO_2 and CH_4 adsorption isotherms onto *β*-zeolite by the Langmuir equation.

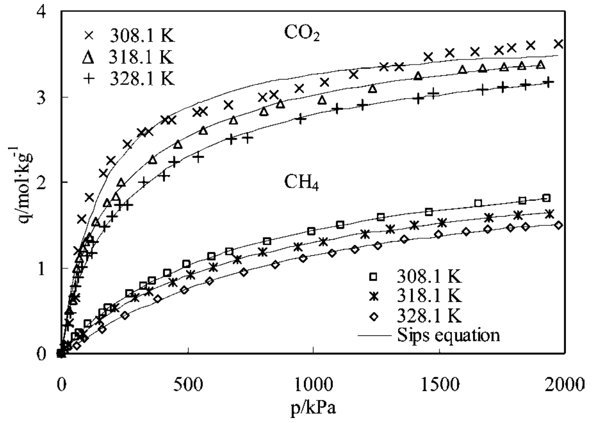

Figure 5. Predicted CO_2 and CH_4 adsorption isotherms onto *β*-zeolite by the Sips equation.

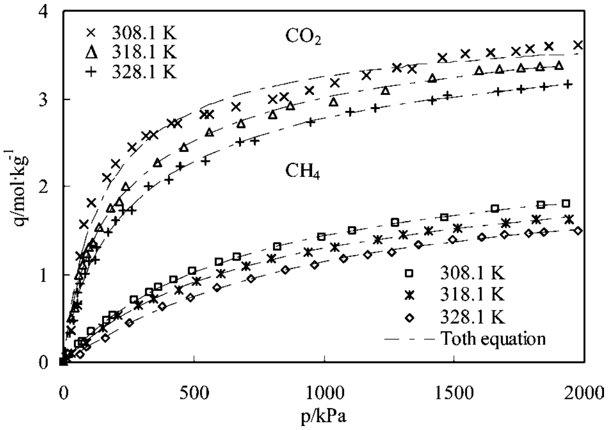

Figure 6. Predicted CO_2 and CH_4 adsorption isotherms onto *β*-zeolite by the Toth equation.

reflected by the distinct adsorbed amounts between CO_2 and CH_4. Since the CO_2 molecule has a high quadrupole moment, thus CO_2 is more polar than the CH_4 molecule and may form stronger interactions with the zeolite surface, and hence more CO_2 than CH_4 could be adsorbed by this adsorbent.

Figures 4 to 6 show the fitted adsorption isotherm curves at various temperatures using the Langmuir, Sips, and Toth equations[17,20−22] along with the experimental data. The Langmuir equation is widely used for physical adsorption from either gas or liquid solution. The expression is obtained from the equilibrium rate expressions of both adsorption and desorption. The mathematical form of this model is

$$q = \frac{q_s bp}{1 + bp} \qquad (2)$$

where q is the amount adsorbate adsorbed, p is the equilibrium pressure, and q_s and b are two isotherm adjustable parameters. Generally, this two-parameter equation can describe monolayer adsorption very well. At low adsorbate pressure, the Langmuir equation reduces to Henry's law that is applicable for describing linear adsorption, as shown below.

$$\lim_{p \to 0}\left(\frac{q}{p}\right) = bq_s = K_p \tag{3}$$

where K_p is the Henry's law constant.

Besides the Langmuir equation, two three-parameter nonlinear equations, that is, the Sips and Toth models, are considered in this work. These two models are widely used, as they can model a great number of sorption data. For multilayer adsorption, the Sips and Toth models are expected to predict the experimental data more closely than the Langmuir equation.

$$\text{Sips: } q = \frac{q_s bp^n}{1 + bp^n} \tag{4}$$

$$\text{Toth: } q = \frac{q_s bp}{(1 + b^n p^n)^{1/n}} \tag{5}$$

where n is an isotherm parameter.

The isotherm parameters are determined numerically by regressing against the experimental isotherm data. In this study, a nonlinear curve-fitting procedure was used to determine q_s, b, and n. The software MATLAB was used to determine the adsorption curve parameters of each adsorption system. The nonlinear least-squares programs were solved using Gauss−Newton methods. In the regression, the objective function used is given as follows:

$$\text{func} = \min \sum_{i}^{N} (q_i^{\text{cal}} - q_i^{\text{exp}})^2 \tag{6}$$

where q_i^{exp} represents the experimental data on the amount adsorbed, q_i^{cal} represents the correlation results, and N is the number of datum points. Moreover, the deviation parameter for the amount adsorbed, AARD (average absolute relative deviation), was used to compare the correlation results with the experimental data:

$$\text{AARD} = \frac{100}{N} \sum_{i}^{N} \left| 1 - \frac{q_i^{\text{cal}}}{q_i^{\text{exp}}} \right| \tag{7}$$

Presented in Tables 3, 4, and 5 are the isotherm parameters obtained from the best fit to the experimental data, along with the AARD calculated according to eq 7. As seen from these tables, all of the correlated isotherms agree very well with the experimental data with an AARD value much less than 5 %, except for the CO_2 308.1 K isotherm where the AARD value is over 16 % for these three equations. Overall, the three models give comparable performance to correlate these adsorption isotherms. It should be noticed that the Langmuir equation has only two parameters and the other two have three parameters. As can be seen from the figures, the Langmuir equation fits the CH_4 adsorption data very well, and the other two equations with one additional adjustable parameter have not led to obviously improved fitting. The modeled results indicate that the CH_4 adsorption equilibria on β-zeolite may be monolayer and dominated by adsorbent−adsorbate interactions rather than lateral adsorbate−adsorbate interactions. The monolayer adsorption for CH_4 with a molecular dimension of less than 0.4 nm[11,12] is reasonable since the maximum number of adsorbed molecules is 1.78 per nm^2 surface area or 2.85 per nm^3 pore volume if estimated from the zeolite structural parameters and the highest loading obtained. Similarly, the CO_2 adsorption may possibly be monolayer as well.

Table 3. Regression Results for the Langmuir Equation

	T(K)	q_s(mol·kg^{-1})	b·10^3(kPa^{-1})	AARD(%)
CO_2	308.1	3.747	6.642	16.13
	318.1	3.618	5.255	4.09
	328.1	3.450	4.340	3.93
CH_4	308.1	2.429	1.466	1.52
	318.1	2.277	1.334	2.06
	328.1	2.336	0.9648	3.43

Table 4. Regression Results for the Sips Equation

	T(K)	q_s(mol·kg^{-1})	b·10^3(kPa^{-1})	n	AARD(%)
CO_2	308.1	3.757	6.864	0.9922	16.36
	318.1	4.074	11.97	0.7920	3.58
	328.1	3.855	8.930	0.8210	4.16
CH_4	308.1	2.520	1.728	0.9623	1.70
	318.1	2.374	1.609	0.9581	1.82
	328.1	2.146	0.6452	1.083	2.52

Table 5. Regression Results for the Toth Equation

	T(K)	q_s(mol·kg^{-1})	b·10^3(kPa^{-1})	n	AARD(%)
CO_2	308.1	3.862	7.273	0.9011	17.31
	318.1	4.321	7.517	0.6536	3.19
	328.1	4.101	5.716	0.6812	3.80
CH_4	308.1	2.610	1.476	0.8951	1.58
	318.1	2.437	1.342	0.9018	1.93
	328.1	2.067	0.9690	1.204	2.78

It can be seen from the equilibrium isotherms that the adsorbent is very selective to carbon dioxide. Preferential adsorption of CO_2 on β-zeolite indicates that this material can be used for the separation of CO_2 from the CO_2 and CH_4 gas mixture. The ideal selectivity of carbon dioxide relative to methane is shown in Figure 7, which decreases as the adsorbate pressure increases and varies from 2 to 7. Compared to the CO_2/CH_4 selectivity obtained at about 1000 kPa for β-zeolite incorporated PES composite membranes,[13] there is still a considerable difference, possibly related to a kinetic mechanism.[7]

The estimation of the isosteric heat of adsorption was calculated from the temperature dependence of the Henry's law constant in accordance with the van't Hoff equation:[20,22]

$$K_p = K_{p0} \exp\left(\frac{-\Delta H}{RT}\right) \tag{8}$$

where K_{p0} is the parameter of the van't Hoff equation and ΔH is the isosteric heat of adsorption at zero loading.

The Henry's law constants are obtained from the linear low-pressure part of the adsorption isotherms, and the results are listed in Table 6. Note that the Henry constants can be agreeably obtained from the Langmuir parameters except for the CO_2 308.1 K isotherm. It is seen that the K_p values at each temperature for CO_2 are much lower than those reported in the literature,[7] whereas for CH_4 these values are comparable to each other. The isosteric heat of adsorption obtained in this work is

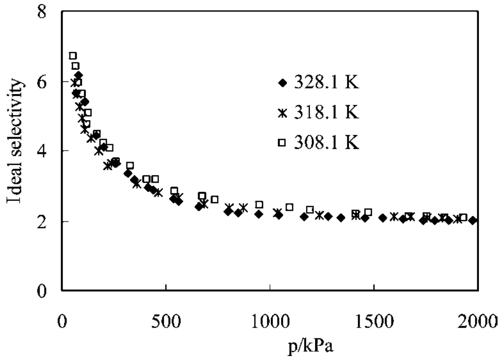

Figure 7. Ideal adsorption selectivity of CO_2 over CH_4 by the β-zeolite.

Table 6. Henry's Law Constants Obtained at Different Temperatures

	308.1 K	318.1 K	328.1 K	
CO_2	2.49	1.90	1.50	this work
$(\cdot 10^2\ mol \cdot kg^{-1} \cdot kPa^{-1})$	9.45	6.19	4.17	7
CH_4	3.56	3.04	2.25	this work
$(\cdot 10^3\ mol \cdot kg^{-1} \cdot kPa^{-1})$	3.17	2.61	2.17	7

21.36 kJ·mol^{-1} for CO_2 and 19.15 kJ·mol^{-1} for CH_4, respectively. These values are different from those reported previously (34.44 kJ·mol^{-1} for CO_2 and 15.86 kJ·mol^{-1} for CH_4).[7] Because of the high affinity of carbon dioxide at low coverage, the estimation of the isosteric heat of adsorption may readily cause obvious differences among different measurement methodologies, explaining the inconsistent isosteric heat value obtained between this work and other methods such as concentration pulse chromatography studies.

Conclusions

This study measured the high-pressure adsorption equilibria of the pure gases CO_2 and methane onto β-zeolite in the pressure range of (0 to 2000) kPa at (308.1, 318.1, and 328.1) K using a static volumetric method. The isothermal data were well-fitted with the Langmuir, Sips, and Toth models. The adsorbent shows very preferential selective adsorption to carbon dioxide, which makes it a very good candidate for carbon dioxide−methane separation.

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