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Adsorption isotherms and ideal selectivities of hydrogen sulfide and carbon dioxide over methane for the Si-CHA zeolite: comparison of carbon dioxide and methane adsorption with the all-silica DD3R zeolite

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Abstract Adsorption isotherms of H_2S, CO_2, and CH_4 on the Si-CHA zeolite were measured over pressure range of 0–190 kPa and temperatures of 298, 323, and 348 K. Acid gases adsorption isotherms on this type of zeolite are reported for the first time. The isotherms follow a typical Type-I shape according to the Brunauer classification. Both Langmuir and Toth isotherms describe well the adsorption isotherms of methane and acid gases over the experimental conditions tested. At room temperature and pressure of 100 kPa, the amount of CO_2 adsorption for Si-CHA zeolite is 29 % greater than that reported elsewhere (van den Bergh et al. J Mem Sci 316:35–45 (2008); Surf Sci Catal 170:1021–1027 (2007)) for the pure silica DD3R zeolite while the amounts of CH_4 adsorption are reasonably the same. Si-CHA zeolite showed high ideal selectivities for acid gases over methane at 100 kPa (6.15 for H_2S and 4.06 for CO_2 at 298 K). Furthermore, H_2S adsorption mechanism was found to be physical, and hence, Si-CHA can be utilized in pressure swing adsorption processes. Due to higher amount of carbon dioxide adsorbed and lower heats of adsorption as well as three dimensional channels of Si-CHA pore structure, this zeolite can remove acid gases from methane in a kinetic based process such as zeolite membrane.

Keywords Adsorption · Zeolite · Pure silica chabazite (Si-CHA) · Acid gas · Natural gas sweetening

List of symbols

b	Affinity parameter of Langmuir or Toth isotherm (kPa^{-1})
b_∞	Affinity parameter of Langmuir or Toth isotherm at infinite temperature (kPa^{-1})
$C_{\mu s}$	Saturation adsorption capacity of adsorbent (mol kg^{-1})
C_μ	Amount adsorbed (mol kg^{-1})
p	Pressure (kPa)
Q^{st}	Isosteric heat of adsorption (kJ mol^{-1})
Q	Isosteric heat of adsorption for Langmuir isotherm in Eq. 3 (kJ mol^{-1})
R	Universal gas constant (kJ K^{-1} mol^{-1})
r	Correlation coefficient
T	Temperature (K)
T_0	Reference temperature (K)
t	Heterogeneity parameter in Toth isotherm
t_0	Heterogeneity parameter at the reference temperature (T_0)
θ	Fractional amount adsorbed
α	Adjustable parameter in Eq. 4

1 Introduction

Removal of acid gases (H_2S, CO_2) impurities from gas streams is of importance for the chemical industries, as industrial processes such as purification of natural gas and sweetening of gas streams in petrochemical plants should be carried out in accordance with strict environmental regulations. Absorption of acid gases into aqueous

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alkanolamine solutions is the most common process employed over the last decades (Gabrielsen et al. 2006; Pacheco and Rochelle 1998; Bolhàr-Nordenkampf et al. 2004; Daviet et al. 1984; Bullin and Polasek 1982; Vrachnos 2006; MacKenzie et al. 1987). However, it is an energy intensive process suffering from some operational problems such as foaming and equipment corrosion.

Acid gas adsorption on solid sorbents and membrane separation processes are of great interest as alternative energy saving processes. Solid sorbents adsorb acid gases physically (physisorption) or chemically (chemisorption) depending on the nature of the solids. Sorbents like zeolites (Jensen et al. 2012; Sircar 1988; Chue et al. 1995; Ishibashi et al. 1999; Cavenati et al. 2004; Ackley et al. 1992, 2003; Delgado et al. 2005; Jayaraman et al. 2004), activated carbon (Sircar and Golden 2000; Fuderer and Rudelstorfer 1976; Sircar 1979; Sircar and Kratz 1988 Sircar et al. 1988; Yang et al. 2011), silica and alumina (Keller et al. 1987), and metal organic frameworks (MOF) (Bae et al. 2009; Saha and Deng 2009; Saha et al. 2010; Xiang et al. 2011) have been utilized for bulk or trace removal of acid gases. Among solid sorbents, zeolites usually adsorb carbon dioxide physically but hydrogen sulfide adsorption on them depends essentially on the polarity of the zeolite framework namely Si/Al ratio (Lutz et al. 1990; Tanada et al. 1982; Cruz et al. 2005; Garcia and Lercher 1992; Karge and Raskó 1987; Yaşyerli et al. 2002; Gaillard et al. 2004; Ferino et al. 1992; Crespo et al. 2008; Bobonich 1995; Herna'ndez-Maldonado et al. 2003). For example, Karge and Raskó (1987) studied the effect of Si/Al on the mechanism of H_2S adsorption for the FAU zeolite. They found that H_2S is adsorbed physically when the Si/Al ratio is greater than 2.5. Furthermore, chemisorption may affect the applicability of the sorbent for pressure swing adsorption (PSA) and even temperature swing adsorption (TSA) processes and make it impossible to use it in a kinetic based process such as membrane separations.

The polarity of zeolite and other polar substances (silica gel and alumina) not only affects the mechanism of H_2S adsorption but also makes H_2O selectively adsorbed over acid gases due to large permanent dipole of the water molecule. Consequently, the presence of even a minor amount of water in the gas streams may significantly decrease the sorption capacity of the sorbent for acid gases. Therefore, various gas adsorption studies have concentrated on pure silica zeolites like silicalite (Dunne et al. 1996) and newly synthesized in fluoride media such as DD3R (Himeno et al. 2007a, b; van den Bergh et al. 2007, 2008; Den Exter 1996; Tomita et al. 2004; Kanezashi et al. 2008; Lin and Kanezashi 2007; Nakayama et al. 2004; Zhu et al. 1999, 2000), Si-CHA (Díaz-Cabañas et al. 1998; Olson et al. 2004; Olson 2002; Hunt et al. 2010a, b) ITQ-3 (Olson et al. 2004; Olson 2002; Camblor et al. 1997) and BEA (Bourrelly et al. 2005; Maurin et al. 2005). These siliceous zeolites can work well in the presence of water because of their hydrophobic nature.

In this paper, the Si-CHA zeolite powder was hydrothermally synthesized and then its phase structure and crystal morphology are characterized by XRD and SEM image, respectively. Adsorption isotherms of CO_2, H_2S, and CH_4 on the Si-CHA zeolite was investigated in the pressure range of 0–190 kPa and temperature range of 298–348 K. Such acid gas adsorption isotherms on Si-CHA zeolite have not been reported elsewhere. Then, adsorption isotherms were regressed well with both Langmuir and Toth equations. Compared with the data reported for the other type of pure silica zeolite, DD3R, high adsorption capacity and selectivity were found for acid gases with the zeolite that was synthesized in this work. Furthermore, H_2S adsorption mechanism was found to be physical that makes it a re-generable sorbent for H_2S adsorption in PSA processes.

2 Experimental

2.1 Synthesis of structure directing agent (SDA): *N*,*N*,*N*-trimethyl-1-adamantylammonium hydroxide

The synthesis procedure that was utilized in this paper is similar to that reported elsewhere (Hunt et al. 2010a, b), except that the alkylating agent (methyl iodide) was substituted with a stronger alkylating agent namely dimethyl sulfate. Fifty grams of 1-adamantylamine (97 %, Aldrich) were dissolved in 500 ml of methanol (>99.9 %, Merck). Fifty-six grams of sodium bicarbonate (99.5 %, Merck) were added to the solution, and then placed in an ice bath while kept under an argon atmosphere. Then, 95 ml of dimethyl sulfate (>99 %, Merck) were added to the solution and the mixture was stirred for 4 days. Methanol was evaporated using a rotary evaporator and the product was extracted from the solids with excess chloroform (>99 %, Merck). The solution was filtered to remove the sodium bicarbonate byproducts and the solids were washed again with excess chloroform. The filtered solution was dried with magnesium sulfate (>98.0 %, Merck). The solution was filtered again to remove the magnesium sulfate. The solution was evaporated using a rotary evaporator, the solids were washed with diethyl ether to dissolve un-reacted 1-adamantylamine and then the resulting solid was placed in a desiccator for 1 day. The crude product was re-crystallized from hot *n*-butanol (>99.5 % Chem-Lab). White crystals of *N*,*N*,*N*-trimethyl-1-adamantylammonium methyl sulfate were obtained after vacuum drying of the obtained solid. The product was

ion-exchanged from its methyl sulfate form to its hydroxide form (TMAdaOH) by dissolving in distilled water while stirring with a strong anion exchange resin for 24 h. Anion exchanging step was repeated with a fresh resin to ensure complete ion exchanging. The solution was then filtered and concentrated to about 0.6 M concentration of the hydroxide form using a rotary evaporator. According to the 1H NMR spectroscopy (Fig. 1), no methyl sulfate anion was detected in the resultant solution which means that methyl sulfate anions were completely substituted by hydroxide anions.

2.2 Synthesis of the Si-CHA zeolite powder

The method for synthesizing pure Si-CHA was first reported by Díaz-Cabañas et al. (1998) that was utilized in this research. 5.91 g of Tetraethylorthosilicate (TEOS: >99 %, Merck) was added to 23.66 ml of a 0.6 M TMAdaOH aqueous solution and then was stirred for 24 h, while covered with parafilm, at room temperature to ensure complete hydrolysis of TEOS. After complete hydrolysis, the mixture was allowed to complete evaporation of ethanol. Finally, the precursor gel with molar composition of 1.0 TEOS/0.5 TMAdaOH/0.5 HF/3.0 H_2O was obtained by adding 0.73 g of HF (38–40 %, Merck). The mixture was mixed and homogenized by hand and then transferred to a 50 ml stainless steel autoclave, heated at 423 K while rotating at 60 rpm. After 40 h synthesis time, solid powder was collected and washed with excess distilled water and then dried. The resulting powder was calcined in air at 850 K to remove fluoride and organic material (SDA).

XRD pattern and SEM image of the synthesized powder were shown in Figs. 2 and 3, respectively. The XRD pattern is the characteristic of pure silica chabazite type, reveals that pure Si-CHA zeolite was obtained.

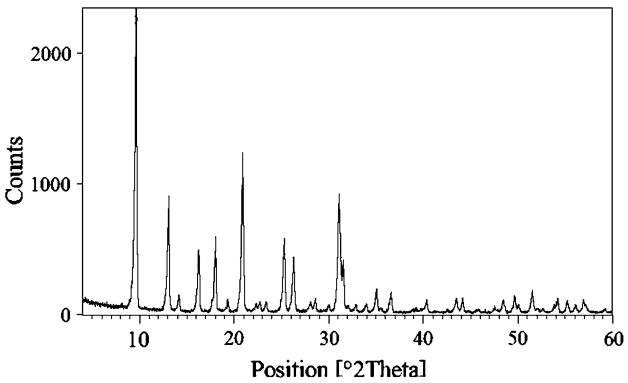

Fig. 2 XRD pattern of calcined Si-CHA powder

2.3 Adsorption apparatus

A simple static-volumetric adsorption apparatus was used to measure the pure gas adsorption isotherms. Figure 4 represents a schematic drawing of the apparatus used. It contains a gas chamber of known volume linked to an adsorption chamber, both immersed in a water bath at constant temperature. One gram of the Si-CHA zeolite was introduced into the adsorption chamber and prior to any adsorption experiments, that chamber was heated at 473 K for 2 h while evacuated with a high vacuum pump. This sample preparation is needed to desorb any adsorbed species. The volumetric method with P–V–T data (NIST 2012) was utilized to calculate the total amount of gas introduced to the gas chamber and that remaining in the system after equilibrium state.

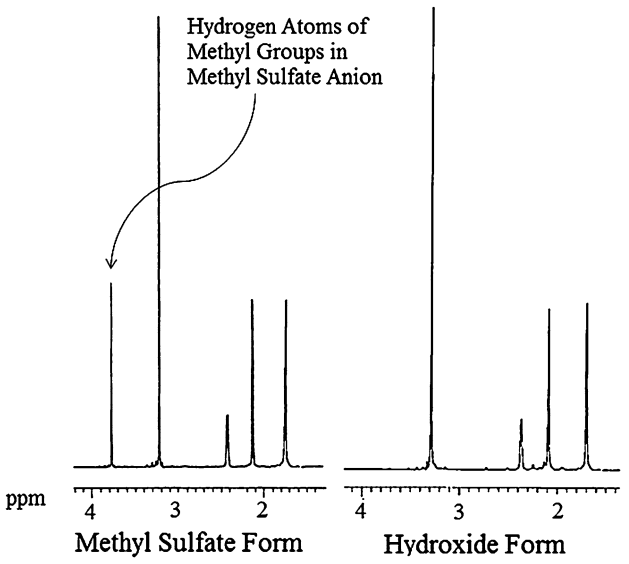

Fig. 1 1H NMR Spectra of *N*,*N*,*N*-trimethyl-1-adamantylammonium methyl sulfate and hydroxide

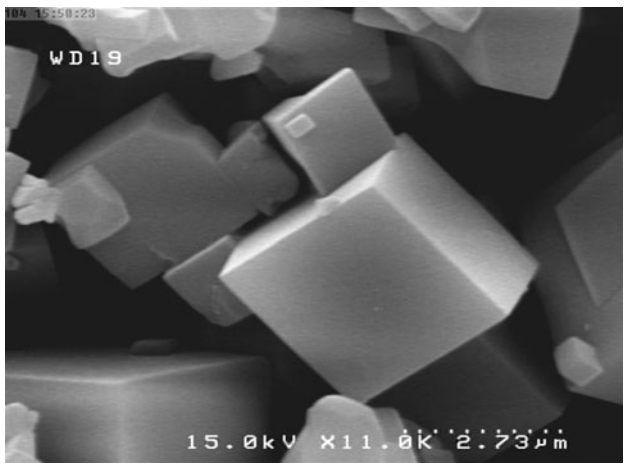

Fig. 3 SEM image of synthesized Si-CHA powder

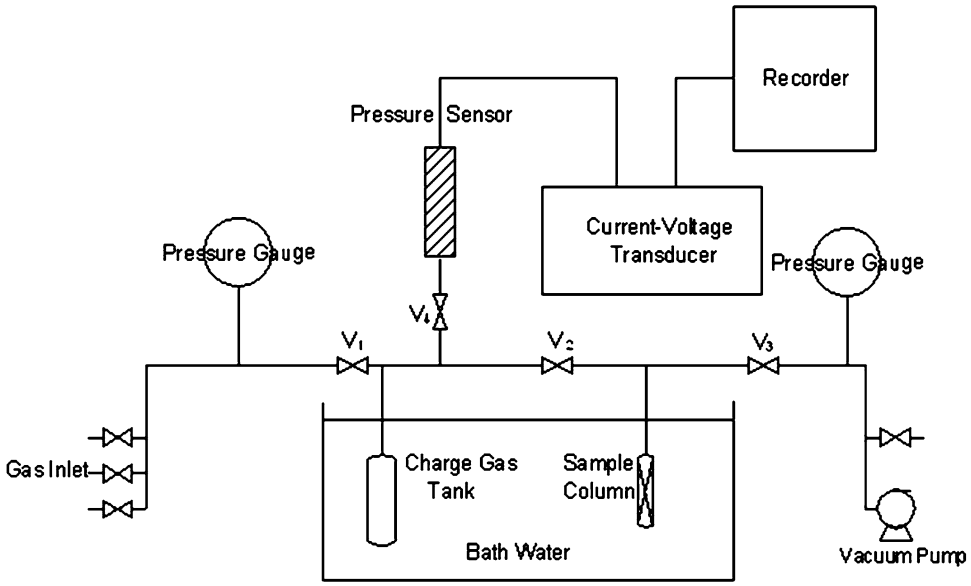

Fig. 4 Schematic diagram of adsorption system

3 Theoretical basis

3.1 Si-CHA zeolite structure and adsorbates molecular dimensions

Si-CHA zeolite is pure silica Chabazite which was first synthesized by Díaz-Cabañas (1998). Si-CHA has the lowest framework density (FD = 15.4) in comparison with the other pure silica zeolites such as DD3R (FD = 17.9) (IZA 2012), ITQ-3 (FD = 16.3), and BEA (FD = 15.6) (Camblor et al. 1999) resulting in the highest micropore volume of 0.30 $cm^3\ g^{-1}$ (DD3R = 0.15, ITQ-3 = 0.23, and BEA = 0.20) and a large BET surface area of 602 $m^2\ g^{-1}$ (Díaz-Cabañas et al. 1998). The more the micropore volume, the more the adsorption capacity that may be achieved.

The crystal structure is built by corner-sharing SiO_4 tetrahedra making two composite building units, large 20-hedra [$4^{12}6^28^6$] cages connected to each other by small octahedra [4^66^2] cages. Connection of 20-hedra cavities through 8-membered rings with an aperture of 3.8 Å × 3.8 Å, forms a three-dimensional channel system, which is accessible to small molecules after removal of SDA molecules by thermal treatment at 850 K.

Kinetic diameter of carbon dioxide and hydrogen sulfide molecules are 3.3 and 3.6 Å, respectively, which are less than 8-membered rings dimension (3.8 Å). Therefore, acid gases can enter the 20-hedra cavities to adsorb on it. In contrast, methane molecule with kinetic diameter of 3.8 Å, close to the opening dimensions, can hardly diffuse into the cavities. So, in the case of acid gases separation from natural gas, the opening dimension gives the CHA zeolite a molecular sieving effect. This effect is much pronounced in kinetic based processes such as membrane separations.

3.2 Adsorption isotherms

Langmuir is the most simple and frequently used model to describe the adsorption on a homogeneous surface. In real situations, the adsorption systems are not homogeneous so that the Langmuir model fails to describe the system with high heterogeneity. One popularly used isotherm equation considering the heterogeneity of the system is the Toth isotherm which is suitable for gas adsorption below capillary condensation region (Duong 1998). Toth equation is commonly used to describe well many data of hydrocarbons and carbon oxides on activated carbon and zeolite:

$$C_\mu = C_{\mu s}\frac{\mathrm{bp}}{(1+(\mathrm{bp})^{\mathrm{t}})^{1/\mathrm{t}}} \quad (1)$$

where C_μ and $C_{\mu s}$ are the amount adsorbed at pressure p and the saturation adsorption capacity of the adsorbent for the gas, respectively. The parameter "b" is called the affinity constant. It is a measure of how strong an adsorbate molecule is attracted onto a surface. In other words, "b" is the gas–solid interaction parameter.

Mathematical form of this equation resembles that of Langmuir equation. The difference between this equation and the Langmuir equation is the additional parameter "t" in the Toth equation. When "t" is unity the equation reduces to the Langmuir equation which is appropriate for ideal surfaces. Hence, the parameter "t" could be considered as the parameter characterizing the system heterogeneity. The source of the system heterogeneity could be the

solid or the adsorbate or a combination of both. If the parameter "t" is deviated further away from unity, the system is said to be more heterogeneous.

3.3 Isosteric heat of adsorption

The isosteric heats of adsorption (Q^{st}) are calculable simply from temperature dependence of the isotherm using the van't Hoff equation.

$$\frac{Q^{st}}{RT^2} = -\left[\frac{\partial \ln p}{\partial T}\right]_{\theta} \tag{2}$$

The isosteric heat of adsorption is independent of surface coverage when the system is ideal and Langmuir model is valid. But, at real adsorption system with some degrees of heterogeneity this heat is a function of coverage. For an adsorption system that Toth equation governs, Q^{st} is calculated by Eq. 5 [Duong 1998]:

$$b = b_{\infty} \exp\left(\frac{Q}{RT}\right) \tag{3}$$

$$t = t_0 + \alpha\left(1 - \frac{T_0}{T}\right) \tag{4}$$

$$Q^{st} = Q - \frac{\alpha R T_0}{t}\left\{\ln\frac{\theta}{(1-\theta^t)^{1/t}} - \frac{\ln\theta}{1-\theta^t}\right\} \tag{5}$$

In which, T_0 is an arbitrary reference temperature, R is universal gas constant, and b_{∞}, Q, t_0 and α are obtained by regression of parameters "b" and "t" at various temperatures.

4 Results and discussion

4.1 Adsorption isotherms

Adsorption isotherms were measured for pure gases, H_2S, CO_2, and CH_4 in the pressure range of 0–190 kPa. These experiments were accomplished at three temperatures (298, 323, and 348 K) for CO_2 and CH_4, and only at 298 K for H_2S (Figs. 5, 6, 7).

The amount of gas adsorption on a solid sorbent depends on the nature of adsorbent–adsorbate interactions. For a non-polar sorbent (like pure silica zeolites that do not have any ion in the crystal structure), the main adsorbent–adsorbate interactions are the dispersion and repulsion forces. These forces are strong functions of the adsorbate molecule polarizability. However, the permanent dipole and quadrupole moments of the molecule can affect this interaction due to induced polarity. Thereby, the adsorbate molecule with high polarizability interacts strongly with the non-polar adsorbent surface and thus it will be highly adsorbed.

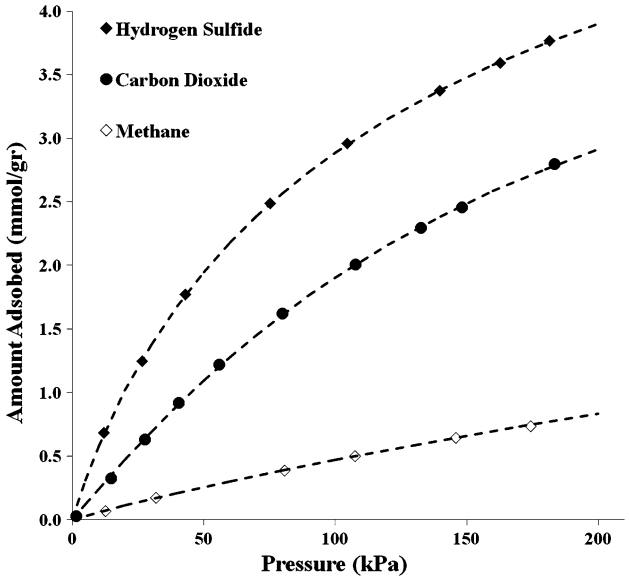

Fig. 5 Adsorption isotherms of sample gases on Si-CHA at 298 K

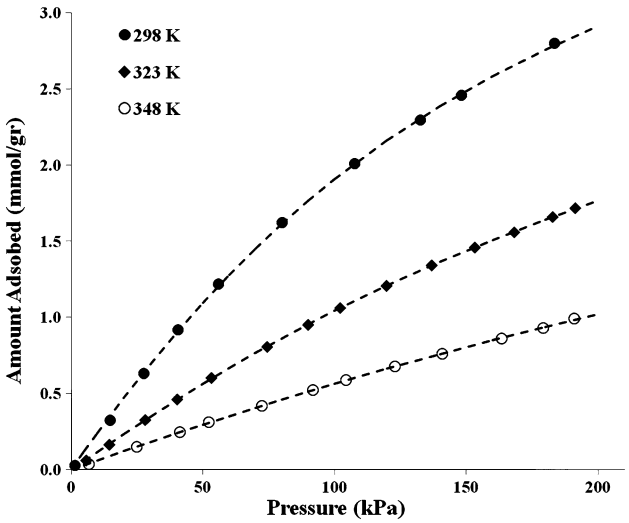

Fig. 6 Adsorption isotherms of carbon dioxide on Si-CHA

The polarizability, permanent dipole and quadrupole moments of sample gases are given in Table 1. H_2S is considered as strongly adsorbed with respect to other gases studied because it has the highest polarizability among them. The obtained order in adsorbed amount is: $H_2S > CO_2 > CH_4$, which is in accordance with the molecule polarizability order.

In comparison with all-silica DD3R zeolite, Si-CHA shows a higher adsorption capacity for CO_2 while the amounts of their CH_4 adsorption are the same. For example, at 298 K and $p = 100$ kPa, CH_4 adsorption on DD3R is about 0.47 mol kg^{-1} (van den Bergh et al. 2007, 2008) that is the same as adsorbed on Si-CHA, 0.47 mol kg^{-1},

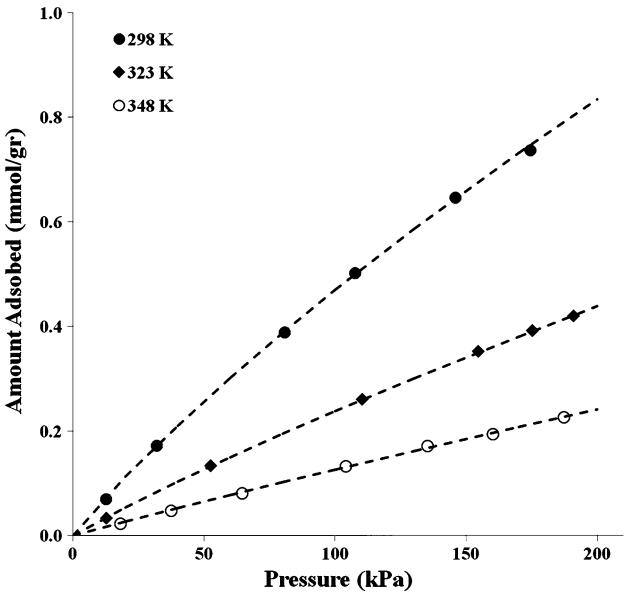

Fig. 7 Adsorption isotherms of methane on Si-CHA

Table 1 Permanent poles and polarizability of sample gases (Lide 2009)

Molecule	Polarizability (10^{-24} cm^2)	Dipole (D)	Quadrupole (10^{-40} cm^2)
H_2S	3.95	0.97833	–
CO_2	2.911	–	-13.4 ± 0.4[a]
CH_4	2.593	–	–

[a] Graham et al. (1989)

reported in this paper. On the other hand, at the same conditions (T = 298 K, $p = 100$ kPa), CO_2 will be adsorbed on Si-CHA zeolite 29 % more than DD3R (1.91 mol kg^{-1} for Si-CHA vs. 1.48 mol kg^{-1} for DD3R (van den Bergh et al. 2007, 2008)).

Isotherms of sample gases at different temperatures were described well by both Langmuir and Toth isotherms with high correlation coefficients ($r > 0.999$). It can be inferred from Table 2, the "b" values of the Si-CHA zeolite have higher affinity to H_2S molecule due to its higher polarizability and lower affinity to CH_4 molecule due to its lower polarizability. The affinity of all molecules decreases with increasing temperature. The value of "t" in Table 2 is less than unity when the adsorbate-adsorbent interaction is stronger than the interactions between adsorbed molecules. But, the "t" parameter values for CO_2 are larger than unity which means that adsorbate–adsorbate interactions may affect the amount adsorbed. However, those interactions are not very high because "t" values are not very larger than one. This impact is further explained in the isosteric heat of adsorption (Sect. 4.3). Another important information taken from Table 2 is that all

Table 2 Toth and Langmuir isotherms parameters ($r > 0.999$)

	Temperature (K)	$C_{\mu s}$ (mol kg^{-1})		b × 10^3 (kPa^{-1})		t
		Toth	Langmuir	Toth	Langmuir	
H_2S	298	7.042	5.72	9.94	10.36	0.780
CO_2	298	4.824	6.803	5.04	3.84	1.363
	323	4.225	6.067	2.83	2.06	1.288
	348	4.704	5.570	1.32	1.13	1.083
CH_4	298	3.927	3.217	1.45	1.71	0.883
	323	2.810	2.472	0.96	1.07	0.936
	348	2.942	1.832	0.45	0.70	0.982

adsorption systems seem to be reasonably homogeneous as the "t" parameter is close to unity especially at higher temperatures for carbon dioxide and methane. Thus, Langmuir isotherm is also suitable isotherm to describe these adsorption systems.

4.2 H_2S adsorption mechanism

After H_2S adsorption experiment, the solid sorbent was evacuated at room temperature. Then, the sample was investigated by means of FTIR spectroscopy to determine the adsorption mechanism of H_2S. When H_2S is chemically adsorbed, infrared spectra reveal bands between 2,500 and 2,600 cm^{-1} which are either assigned to the SH stretching vibration of SH^- groups for dissociative adsorption or to the HSH stretching vibrations for non-dissociative adsorption. The infrared spectra of the utilized sample (Fig. 8) show no band in the aforementioned range

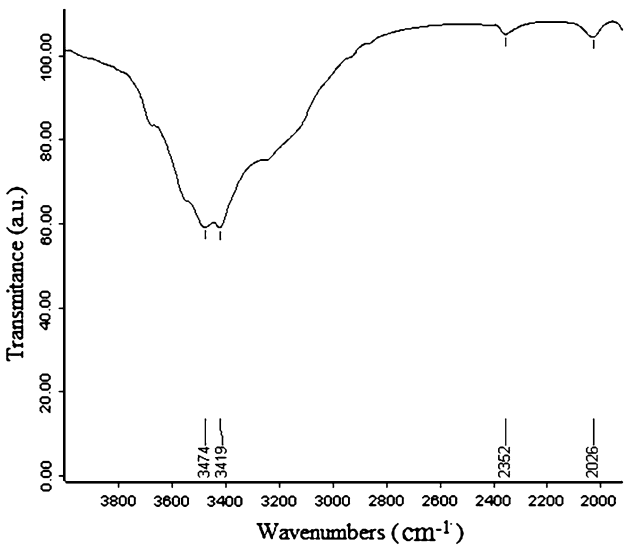

Fig. 8 FTIR spectra of the Si-CHA powder after H_2S adsorption, followed by evacuation at room temperature

(2,500–2,600 cm^{-1}). Therefore, it can be inferred that Si-CHA is a physical sorbent for H_2S.

4.3 Isosteric heats of adsorption and ideal selectivities at 100 kPa

The stronger the interaction of the molecule with the wall of the zeolite, the higher the adsorption energy, and hence, the higher the isosteric heat of adsorption. Calculation of isosteric heat of adsorption is of prime concern as it describes the affinity of the zeolite to the molecule.

Isosteric heats of adsorption for carbon dioxide and methane are derived from Eq. 5 for two isotherms, Langmuir and Toth, and depicted versus loading (mol kg^{-1}) in Fig. 9. Calculated adsorption heats are almost constant at all loadings for the two isotherms but they are somewhat greater for Toth model. This figure shows that carbon dioxide interacts with the Si-CHA walls strongly in comparison with methane. This is attributed to the higher polarizability of carbon dioxide molecule. Carbon dioxide has a permanent quadrupole moment that can affect the amount adsorbed. Thus, molecule–molecule interactions may influence the isosteric heat of adsorption when carbon dioxide loading increases. Therefore, isosteric heat of adsorption for carbon dioxide increases very slightly with increase in loading: from 21.0 kJ mol^{-1} at zero coverage to 21.1 kJ mol^{-1} at full surface coverage condition. The same trend in isosteric heat of adsorption was found for carbon dioxide adsorption on pure silica DD3R zeolite when the loading is greater than 0.5 mol kg^{-1} (Himeno et al. 2007a). In the case of methane, isosteric heat of adsorption tends to decrease with increasing adsorption at low loadings. The higher heat at low loadings may be ascribed to the external hydroxyl groups interacting with methane molecules (Den Exter 1996). Plateau behavior in

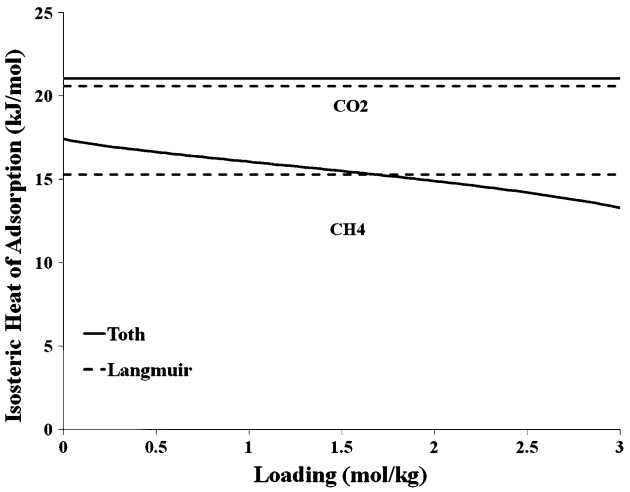

Fig. 9 Isosteric heat of adsorption for CO_2 and CH_4

Table 3 Ideal selectivities at 100 kPa

	H_2S/CH_4	CO_2/CH_4		
Temperature (K)	298	298	323	348
Ideal selectivity	6.15	4.06	4.38	4.47

Fig. 9 demonstrates that the Si-CHA zeolite is energetically uniform, at least toward the carbon dioxide and methane molecules.

According to the calculated adsorption heats by Eq. 5, the limit of isosteric heats of adsorption at zero coverage for carbon dioxide and methane are 21.0 and 17.1 kJ mol^{-1}, respectively. In comparison, they are lower than those reported for the other pure silica zeolite, DD3R, 32 and 18.2 kJ mol^{-1}, respectively (Himeno et al. 2007a, b). This means that carbon dioxide molecules are weakly adsorbed to the Si-CHA surfaces, compared to the DD3R zeolite. Loosely connected molecules can diffuse faster. Thus, it is expected that diffusion of carbon dioxide molecule through the intra-crystalline pores of Si-CHA to be higher than that for DD3R. Therefore, lower heat of adsorption and higher amount of adsorption for CO_2 as well as three dimensional pore structure of Si-CHA make it an effective sorbent for selectively removing carbon dioxide over methane especially in the kinetic based separations such as membrane processes in which diffusivity of molecule is important.

Ideal selectivities of acid gases at 100 kPa are presented in Table 3. According to this table, ideal selectivities of hydrogen sulfide and carbon dioxide are 6.15 and 4.06–4.47, respectively, indicating that Si-CHA can selectively adsorb acid gases over methane. In comparison, the pure silica zeolite, DD3R, shows an ideal selectivity of about 3.15 for CO_2 at the same conditions (extracted from experimental data reported in the literature van den Bergh et al. (2007, 2008)). Thus, CO_2/CH_4 ideal selectivity of Si-CHA at those conditions is almost 30 % higher than that of DD3R. Furthermore, with increasing temperature, CO_2/CH_4 selectivity improves slightly; however, adsorption amounts of all gases decrease.

5 Conclusion

Adsorption isotherms of methane and CO_2 and H_2S on the Si-CHA zeolite were measured accurately over pressure range of 0–190 kPa and temperatures 298, 323, and 348 K. The isotherms follow a typical Type-I shape according to the Brunauer classification. Both Langmuir and Toth isotherms describe well the adsorption isotherms of methane and acid gases over experimental conditions tested. Plateau behavior of isosteric heats of adsorption versus adsorbate

loading indicates that the adsorption system is energetically homogeneous. Si-CHA zeolite showed high adsorption selectivity for acid gases over methane. CO_2/CH_4 ideal selectivity of Si-CHA at 100 kPa and 298 K is almost 30 % higher than that of the DD3R zeolite. Due to higher amount adsorbed and lower heats of adsorption for carbon dioxide as well as three dimensional channels of Si-CHA pore structure, this zeolite may be a good candidate for removing acid gases from methane through a zeolite membrane.

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