



## Effect of Particle Size of NaX Zeolite on Adsorption of CO<sub>2</sub>/CH<sub>4</sub>

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### ABSTRACT

In the present work, nano-NaX zeolite and micro-NaX zeolite were synthesized via hydrothermal method. Then, the adsorption capacities and isotherms of pure gases CO<sub>2</sub> and CH<sub>4</sub> on the synthesized zeolite nanoparticles were determined at three temperatures of 288, 298 and 308 K and various pressures from 1 up to 20 bar. Adsorption capacities of CO<sub>2</sub> on the nano-sized zeolites NaX were higher than CH<sub>4</sub>. The selectivity of CO<sub>2</sub>/CH<sub>4</sub> of the nano-sized zeolites NaX was 5.47 at 288 K and pressure of about 20 bar. The results of the experimental data followed the Langmuir-Freundlich adsorption isotherm. Reduction of the particle size from micrometer to nanometer resulted in increasing the adsorption capacity for carbon dioxide on the X zeolite nanoparticles about 28% (from 5.067 to 6.536 mmol/g) at 288 K and 20 bar.

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## 1. INTRODUCTION

Separation of carbon dioxide from methane is an important process in natural gas purification. CO<sub>2</sub> in the presence of water is a corrosive gas and decreases the heat content of natural gas [1-4]. So, the need for CO<sub>2</sub> capture is of great importance. Several methods exist for separation of CO<sub>2</sub>/CH<sub>4</sub> mixture such as absorption using solvents, membrane technology, cryogenic process, adsorption and etc. [5-8]. Among them, the separation of carbon dioxide from natural gas by adsorption process could be an economic and easy approach in industrial applications [2, 4, 5]. The adsorbents such as zeolites, activated carbons [6, 9, 10] and hydrotalcite [11] have been widely examined for CO<sub>2</sub> capture [7]. Among them, zeolites have attracted attention due to its extensive use as adsorbents for CO<sub>2</sub> capture and relatively low cost [2, 4]. Zeolites are the most important family in microporous materials with uniform and regular pores in molecular dimensions, that consists of [AlO<sub>4</sub>]<sup>5-</sup> and [SiO<sub>4</sub>]<sup>4-</sup> tetrahedra [12-14]. They have been widely used in the petrochemical industry and oil-

refining as catalysts and adsorbents because of their well-defined structure, good shape-selectivity and large pore volume [12, 13, 15]. Microporous zeolitic materials such as zeolite X exhibit high adsorption and separation capacities for CO<sub>2</sub> capture [2].

There are large number of research papers about the synthesis of micro-zeolites and nano-zeolites by various methods and from various precursor materials in the presence of organic templates and template-free condition under different conditions [13, 14, 16-19]. Reduction of the particle size from micrometer to nanometer plays a major role in material characteristics and their applications in adsorption and catalysis [17, 20]. Recently, the synthesis of nanometer-sized zeolites has been increased due to their different activities and large external surface area compared to micrometer-sized zeolite crystals [16, 17, 19, 21]. The synthesis of various nanometer-sized zeolites have been reported in the presence of organic templates and without the assistance of any organic additives [15-17, 22-24].

In recent years, a large number of works have been reported about the separation of CO<sub>2</sub>/CH<sub>4</sub> using various materials, e.g. zeolites [1, 4, 25], etc in the micron size range. But, there is limited research on the application

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of zeolite nanoparticles for the separation of gases. For example Jiang et al. [3] have reported that the synthesized T-type zeolite nanoparticles showed adsorption capacities higher than micro-level T-type zeolite for the separation of CO<sub>2</sub>/N<sub>2</sub> and CO<sub>2</sub>/CH<sub>4</sub>. Cheung et al. [26] pointed that the synthesized nano-sized zeolite NaKA exhibited comparable CO<sub>2</sub> adsorption properties to the large zeolite NaKA crystals.

In this study, the nano-NaX zeolite and micro-NaX zeolite were prepared by hydrothermal method, then nanocrystalline NaX zeolite was compared with micro-zeolite NaX by X-ray diffraction (XRD) and Brunauer–Emmet–Teller (BET) analyses and examined as solid sorbents for CO<sub>2</sub>/CH<sub>4</sub> separation. At the end, the performance of nano-NaX zeolite in the adsorption was compared to the synthesized zeolite with large particle size (micro-zeolite NaX) and the adsorption capacities of CO<sub>2</sub> and CH<sub>4</sub> were determined by volumetric measurements.

## 2. EXPERIMENTAL

**2. 1. Materials** All reactants were used as received without any further purification. The chemical reagents used contained fumed silica (Sigma Aldrich), NaOH (Merck) and NaAlO<sub>2</sub> (Sigma Aldrich) were employed in zeolite synthesis. Methane (99.999%), helium (99.999%) and carbon dioxide (99.99%) were obtained from gas, Iran for the adsorption isotherm measurements.

**2. 2. Synthesis of Micro and nano-NaX zeolites** Micrometer NaX zeolite (micro-NaX) and Nanometer NaX zeolite (nano-NaX) were synthesized for the comparison of CO<sub>2</sub>, CH<sub>4</sub> adsorption.

The micro and nano-NaX zeolites were prepared using established procedures by the hydrothermal synthetic method [16, 17, 19, 27, 28]. Aluminosilicate gel with molar ratios of 5.5 Na<sub>2</sub>O:1.0 Al<sub>2</sub>O<sub>3</sub>:4.0 SiO<sub>2</sub>:190 H<sub>2</sub>O was utilized for synthesis of samples. The hydrogel was made by mixing specific amounts of sodium aluminate and sodium hydroxide pellets in deionized water, followed by adding of silica source. The mixture was stirred for 24 h, then to prepare nano-NaX zeolite, the gel was made at 333 K for 2 days in a shaker for hydrothermal crystallization. The powder product was separated via centrifugation, washed several times with DI water, and then dried at 423 K for 5 h. In other synthesis to prepare larger particle sizes of NaX-micro zeolite, the gel was synthesized with the same aluminosilicate gel composition; however the hydrothermal crystallization was done at 363 K in 48 h under autogenous pressure with no shaking.

**2. 3. Characterization** The solid products were characterized by a variety of routine techniques. X-Ray

diffraction (XRD) spectra of the samples were taken by Philips 1830 diffractometer with Cu-K $\alpha$  radiation. XRD was used to characterize the crystal phases of synthesized materials. The XRD data were aggregated in the 2 $\theta$  range between 5° and 40° with a 2 $\theta$  step size of 0.02° and a step time of 1 s. The nitrogen adsorption-desorption experiments were measured on a Micromeritics model ASAP 2020 sorption analyzer at liquid nitrogen temperature (77 K). Before measurement, the samples were degassed at 473 K for 2 h. The specific surface area (S<sub>BET</sub>) was determined from the linear part of the isotherm using Brunauer–Emmet–Teller (BET) equation.

**2. 4. CO<sub>2</sub> Adsorption Measurement** The CO<sub>2</sub> and CH<sub>4</sub> adsorption capacities of the products were measured using a standard system based on volumetric method. The adsorption capacities of the micro- and nano-NaX zeolites was determined using the setup shown in Figure 1. First, 1 g of a sample was loaded into the adsorption cell. Before measurement, the samples were degassed by using the vacuum pump at 523 K for 1.5 h. The adsorption measurements of gases were carried out using high purity carbon dioxide (99.999%), methane (99.99%) and helium as the purge gas. The system after degassing was cooled to ambient temperature. The equilibrium adsorption experiments were carried out under different temperatures (288, 298 and 308 K) and pressure ranging of 1-20 bar. The adsorption process was carried out by opening the valves 3, 4, 5, 7, 8 and closing other valves; to reach a pressure balance in the reference cell valves 7 and 8 were opened. Then, by opening valve 10 the pressure decreased. The pressure of adsorption cell decreased because of some gas adsorption and some dead volume. The dead volume includes void volume and volume of the connection tubes. The dead volumes were measured via helium tests and pressure decrease calculated the exact gas adsorption. Details of the procedures and the equipments applied are explained in literature [29, 30].

After estimation of adsorption capacity of CO<sub>2</sub> and CH<sub>4</sub>, the sorption selectivity (S) of the adsorbent was obtained according to the following formula (Equation (1)) [4, 31, 32]:

$$S_{\text{CO}_2/\text{CH}_4} = \frac{q_{\text{CO}_2}/P_{\text{CO}_2}}{q_{\text{CH}_4}/P_{\text{CH}_4}} \quad (1)$$

where q is the equilibrium adsorbed amount at the equilibrium pressure for each gas.

## 3. RESULTS AND DISCUSSION

**3. 1. Properties of Adsorbents** The X-ray diffraction technique is a powerful tool for characterization and identification of crystalline materials as zeolites [19, 33].

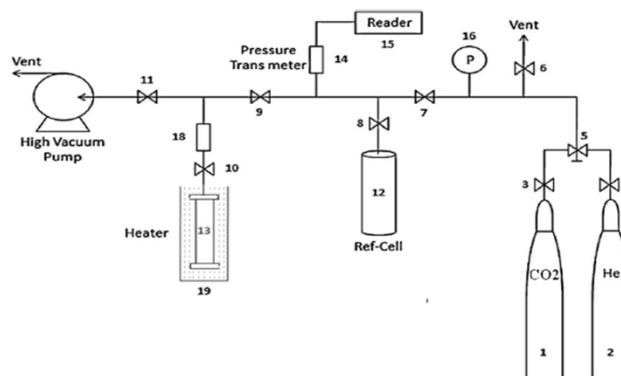


Figure 1. Schematic diagram for experimental set up for adsorption capacity test

TABLE 1. Characteristics of nano-NaX and micro-NaX.

Sample	BET Surface area ( $\text{m}^2\text{g}^{-1}$ )	External surface area ( $\text{m}^2\text{g}^{-1}$ )	Crystal size by XRD (nm)	Micropore volume ( $\text{cm}^3\text{g}^{-1}$ )
Nano-NaX	580	120	30	0.25
Micro-NaX	520	45	700	0.18

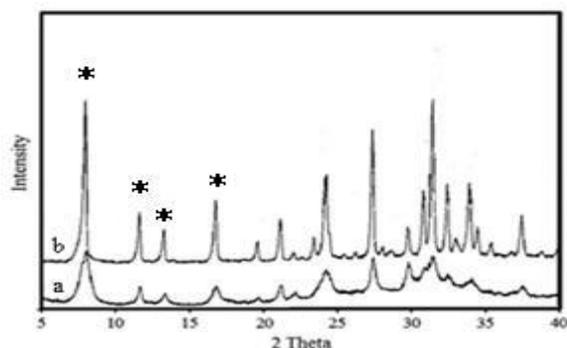


Figure 2. X-ray diffraction patterns of samples (a) nano-NaX and (b) micro-NaX, \* NaX.

The XRD patterns of the synthesized samples are shown in Figure 2 to compare the nano-sized zeolite NaX with micro-NaX zeolite. It is clear from Figure 2 that characteristic XRD peaks of samples at  $2\theta$  are exactly compatible with the reference sample and in the both samples NaX zeolite phase is appeared. It can be observed that the diffraction peak widths and intensities are different. Micro-NaX has high intensity and very sharp peaks; however the diffraction lines of the nano-NaX are much broader with less intensity because the crystal sizes of nano-NaX are smaller than that of micro-NaX.

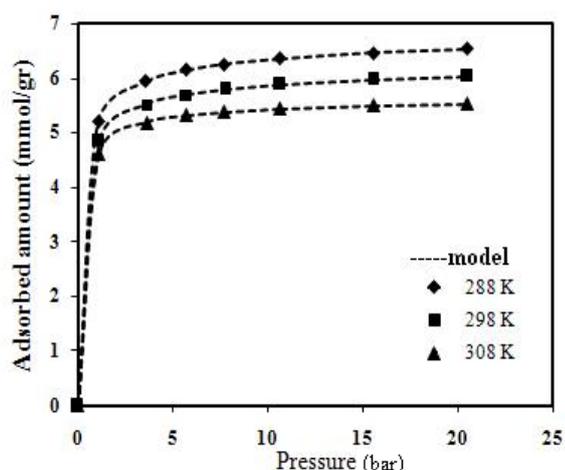
Therefore, lower crystallization temperatures and strong shaking conditions prepare the small crystal sizes. The average crystal size of the prepared zeolites was calculated using Scherrer's equation. The BET analysis was used to determine the external surface area,

micropore volume and surface area. The BET results for micro-NaX and nano-NaX are evaluated and compared with each other in Table 1. As can be seen from this table, the decrease in particle size from micro-NaX to nano-NaX concludes to larger surface area of nano-NaX in comparison with the micro-NaX. Also, nano-NaX has smaller crystal size and higher external surface area.

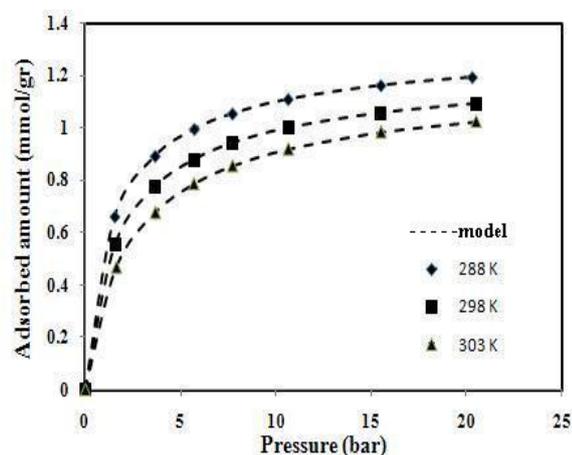
### 3. 2. Adsorption Process

**3. 2. 1. Adsorption Performance** The  $\text{CO}_2$  and  $\text{CH}_4$  adsorption isotherms of the synthesized zeolite nanoparticle were obtained at temperatures of 288, 298 and 308 K for pressures variable in range of 0 to 20 bar by volumetric method. The isotherms are shown in Figures 3 and 4. The  $\text{CO}_2$  and  $\text{CH}_4$  adsorption isotherms at different temperatures exhibited a type I isotherm according to the IUPAC classification. It can be seen that with increase of temperature at a given pressure, the amounts of adsorbed gases ( $\text{CO}_2$ ,  $\text{CH}_4$ ) decreased on the sample nano-NaX, as the adsorption is physisorption in micropores [5]. On the other hand, as temperature increases, the adsorbed molecules achieve sufficient energy to desorb from the surface of zeolite [3].

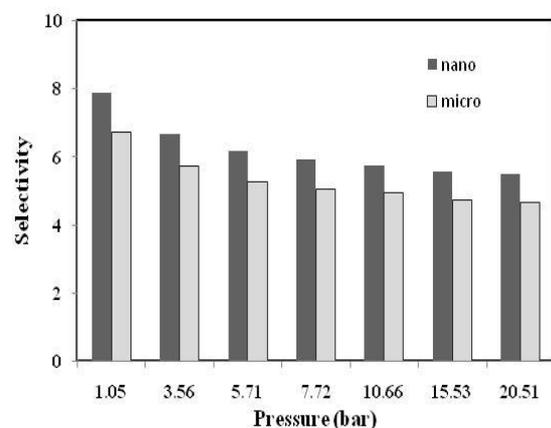
Comparing  $\text{CO}_2$  and  $\text{CH}_4$  isotherms of nano-NaX exhibit that the amount adsorbed increased with increasing pressure, however the slope of adsorption is very sharp at low pressures. The high  $\text{CO}_2$  uptake by zeolite can be attributed to the interaction of  $\text{CO}_2$  with polar zeolite X and smaller molecular size of  $\text{CO}_2$  compared to zeolite NaX [4], whereas  $\text{CH}_4$  only at high pressures shows weak interaction with the adsorbent surface by creating induced polarity and London forces.



**Figure 3.** Adsorption isotherms of CO<sub>2</sub> on the nano-NaX zeolite at different temperatures



**Figure 4.** Adsorption isotherms of CH<sub>4</sub> on the nano-NaX zeolite at different temperatures



**Figure 5.** Selectivity of CO<sub>2</sub>/CH<sub>4</sub> for nano and micro zeolite at different pressures and fixed temperature of 288 K

According to the CO<sub>2</sub> and CH<sub>4</sub> isotherms, the adsorption capacities of nano-NaX and micro-NaX zeolites are presented in Table 2. It can be seen that the amounts of CO<sub>2</sub> and CH<sub>4</sub> adsorbed on the nano-zeolite sample were higher than micro-zeolite. The CO<sub>2</sub> adsorption capacity of nano-NaX was 6.536 mmol/g that was about 28% higher than the CO<sub>2</sub> adsorption capacity of microzeolite at 288K. The increase of surface area or micropore volume due to reduction of the particle size from micrometer to nanometer could be the reason for the enhancement of CO<sub>2</sub> adsorption capacity in nano-NaX. Therefore, the nano-NaX showed higher CO<sub>2</sub> adsorption capacity than the micro-NaX. Also, the adsorption capacity of CO<sub>2</sub> was higher than CH<sub>4</sub>, which can be ascribed to that CO<sub>2</sub> has higher polarizability than CH<sub>4</sub> [4].

**3. 2. 2. Selectivity of CO<sub>2</sub>** To estimate the selectivity of CO<sub>2</sub>-over-CH<sub>4</sub>, we calculated the ratios of the amount of CO<sub>2</sub> to CH<sub>4</sub> adsorbed on nano-NaX and micro-NaX using Equation (1) and the results are presented in Table 2. These ratios showed that the nano-NaX had higher selectivity than micro-NaX. The CO<sub>2</sub>/CH<sub>4</sub> selectivity of nano-NaX at temperature of 288 K and about 20 bar is 5.47, that is much higher than the selectivity of micro-NaX. This can be explained that micropores increase the adsorption surface as reduction of the particle size from micrometer to nanometer occurs. It can be seen in Table 1 that nano-NaX has the micropore volume larger than micro-NaX. Furthermore, the selectivity of CO<sub>2</sub> over CH<sub>4</sub>, at temperature 288 K was calculated using Equation (1) for each zeolite and the results are shown in Figure 5. As shown in Figure 5, the CO<sub>2</sub>/CH<sub>4</sub> selectivity of nano-NaX is greater than micro-NaX.

Table 3 presents the selectivity of CO<sub>2</sub> over CH<sub>4</sub> and the amounts of CO<sub>2</sub> and CH<sub>4</sub> adsorbed at the experimental conditions for the nano-sized zeolite NaX. The data in Table 3 indicated, when the adsorption temperature increased, the equilibrium selectivity of CO<sub>2</sub>/CH<sub>4</sub> increased.

The effects of temperature and pressure on the selectivity of CO<sub>2</sub> over CH<sub>4</sub> using nano-NaX are shown in Figure 6. It is clear from figure that the selectivity of CO<sub>2</sub>/CH<sub>4</sub> reduces with the increase of pressure and comes near to a constant value. This can be explained by the fact that adsorption amount of CO<sub>2</sub> at low pressure is high. Furthermore, the selectivity of CO<sub>2</sub> over CH<sub>4</sub> increases with the increase of temperature, which may be ascribed to temperature that has the stronger effect for desorption of CH<sub>4</sub> from the surface compared to CO<sub>2</sub> [4].

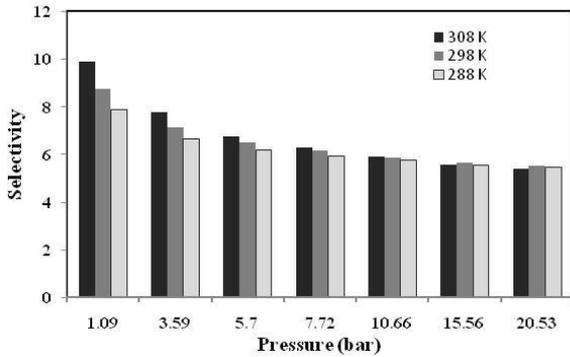
Although the higher selectivity of CO<sub>2</sub>/CH<sub>4</sub> obtained at higher temperatures (at 308 K), but the adsorption process for higher adsorptive capacity and saving energy is preferred to work at lower temperatures.

**TABLE 2.** Adsorption capacities of CO<sub>2</sub> and CH<sub>4</sub> for micro-NaX and nano-NaX (288 K, 20 bar)

Sample	q (CO <sub>2</sub> , mmol/g)	q (CH <sub>4</sub> , mmol/g)	Equilibrium selectivity (CO <sub>2</sub> /CH <sub>4</sub> )
micro-NaX	5.067	1.09	4.65
nano-NaX	6.536	1.194	5.47

**TABLE 3.** Adsorption selectivity of CO<sub>2</sub>/CH<sub>4</sub> over the zeolite nanoparticles at temperatures 288, 298 and 308 K

	Pressure	Temperature (K)	q (mmol/g)		Selectivity nano
			CO <sub>2</sub>	CH <sub>4</sub>	
1	1.05	288	5.201	0.661	7.87
	3.56		5.937	0.892	6.66
	5.71		6.146	0.995	6.18
	7.72		6.259	1.055	5.93
	10.66		6.364	1.109	5.74
	15.53		6.469	1.163	5.56
	20.51		6.536	1.194	5.47
2	1.08	298	4.853	0.554	8.76
	3.59		5.52	0.774	7.13
	5.7		5.696	0.879	6.48
	7.73		5.795	0.942	6.15
	10.65		5.884	1.000	5.88
	15.53		5.973	1.057	5.65
	20.53		6.027	1.093	5.51
3	1.09	308	4.621	0.467	9.89
	3.59		5.184	0.677	7.77
	5.7		5.315	0.785	6.77
	7.72		5.381	0.854	6.30
	10.66		5.438	0.919	5.92
	15.56		5.490	0.984	5.58
	20.53		5.520	1.023	5.39

**Figure 6.** Selectivity of CO<sub>2</sub>/CH<sub>4</sub> for nano zeolite sample at different pressures and temperatures

**3. 2. 3. Adsorption Model** Langmuir–Freundlich (L–F) equation was used to fit the adsorption data of gases on the zeolite nanoparticle for describing of CO<sub>2</sub> and CH<sub>4</sub> adsorption and equation is expressed by Equation (2), as [3, 4]:

$$q = q_m \frac{(bP)^{1/n}}{1 + (bP)^{1/n}} \quad (2)$$

where  $q$  (mmol/gr) is the amount of gas adsorbed on the zeolite at equilibrium pressure of  $P$  (bar). The

parameters  $q_m$ ,  $b$  and  $n$  are the maximum capacity of adsorption, the affinity constant for adsorption and the heterogeneity parameter of system, respectively.

The square of residuals (SOR) of the experimental and calculated adsorption capacity (mmol/g) were determined by Equation (3) [3], as follow:

$$\text{SOR} = \sum_{i=1}^n (q_{i,\text{exp}} - q_{i,\text{cal}})^2 \quad (3)$$

where  $q_{i,\text{exp}}$  and  $q_{i,\text{cal}}$  are the experimental and calculated adsorption capacity (mmol/g) at (T,P) given, respectively.

The dashed lines in Figures 3 and 4 show the results of fitting by the Langmuir–Freundlich model equation which fit the experimental data well. The values of Langmuir–Freundlich equation parameters are summarized in Table 4. Comparison of  $n$  values for CO<sub>2</sub> against CH<sub>4</sub> in Table 4 indicates that CO<sub>2</sub>/zeolite X system is more heterogeneous [3], because the deviation of  $n$  from unity for CO<sub>2</sub> is more considerable. With the increase of adsorption temperature, the parameter  $b$  decreases which shows that at lower temperature, affinity of the adsorbate to the zeolite surface is stronger. Also, the parameter  $q_m$  becomes lower with the increase of temperature, suggesting that the amount adsorbed decreases at higher temperature.

The data in Table 4 also indicated, all parameters for CO<sub>2</sub> are higher than CH<sub>4</sub> at the same conditions.

**TABLE 4.** Parameters of Langmuir–Freundlich equation for CO<sub>2</sub> and CH<sub>4</sub> at 288, 298 and 308 K

Component	T(K)	q <sub>m</sub> (mmol/g)	b *10 <sup>2</sup> (kPa) <sup>-1</sup>	n
CO <sub>2</sub>	288	6.972	6.790	1.822
CH <sub>4</sub>		1.332	0.630	1.189
CO <sub>2</sub>	298	6.356	6.745	1.694
CH <sub>4</sub>		1.245	0.489	1.173
CO <sub>2</sub>	308	5.652	6.590	1.316
CH <sub>4</sub>		1.201	0.364	1.150

It could be explained that nano-NaX shows more tendency for CO<sub>2</sub> against CH<sub>4</sub> as carbon dioxide has higher polarisability than CH<sub>4</sub>.

#### 4. CONCLUSIONS

The nano-sized zeolite NaX (nano-NaX) and the zeolite of larger particle size (micro-NaX) were synthesized via the hydrothermal method, then they were tested comparatively for CO<sub>2</sub> and CH<sub>4</sub> adsorption. The nano-sized zeolite NaX showed promising performance for capture of CO<sub>2</sub> from CO<sub>2</sub>/CH<sub>4</sub> mixture. Experimental adsorption isotherms of CO<sub>2</sub> and CH<sub>4</sub> on the nano-sized zeolite NaX were measured by the volumetric method at 288, 298 and 308 K with pressures from 0 to 20 bar. At 288 K and about 20 bar, the adsorption capacity of the nano-sized zeolite NaX for CO<sub>2</sub> was 6.536 mmol/g, that was about 28% higher than micro-NaX. The nano-sized zeolite NaX showed high selectivity for CO<sub>2</sub> over CH<sub>4</sub>. Also, the nano-sized zeolite NaX exhibited higher selectivity compared to the zeolite of larger particle size.

#### 5. ACKNOWLEDGEMENTS

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#### 6. REFERENCES

- Li, Y., Yi, H., Tang, X., Li, F., and Yuan, Q., "Adsorption separation of CO<sub>2</sub>/CH<sub>4</sub> gas mixture on the commercial zeolites at atmospheric pressure". *Chemical Engineering Journal*, Vol. 229, (2013), 50-56.
- Yu, L., Gong, J., Zeng, C., and Zhang, L., "Synthesis of Binderless Zeolite X Microspheres and Their CO<sub>2</sub> Adsorption Properties". *Separation and Purification Technology*, Vol. 118, (2013), 188-195.
- Jiang, Q., Rentschler, J., Sethia, G., Weinman, S., Perrone, R., and Liu, K., "Synthesis of T-type zeolite nanoparticles for the separation of CO<sub>2</sub>/N<sub>2</sub> and CO<sub>2</sub>/CH<sub>4</sub> by adsorption process". *Chemical Engineering Journal*, Vol. 230, (2013), 380-388.
- DoroudianRada, M., Fatemi, S., and Mirfendereskic, S.M., "Development of T type zeolite for separation of CO<sub>2</sub> from CH<sub>4</sub> in adsorption processes". *Chemical Engineering Research and Design*, Vol. 90, (2010), 1687-1695.
- Zhang, Z., Zhang, W., Chen, X., Xia, Q., and Li, Z., "Adsorption of CO<sub>2</sub> on zeolite 13X and activated carbon with higher surface area". *Separation Science and Technology*, Vol. 45, (2010), 710-719.
- Chen, C., Park, D.W., and Ahn, W.S., "CO<sub>2</sub> capture using zeolite 13X prepared from bentonite". *Applied Surface Science*, Vol. 292, (2013), 63-67.
- Anbia, M. and Hoseini, V., "Development of MWCNT@MIL-101 hybrid composite with enhanced adsorption capacity for carbon dioxide". *Chemical Engineering Journal*, Vol. 191, (2012), 326-330.
- Cejka, J., Bekkum, H.V., Corma, A., and Schuth, F., "Introduction to Zeolite Science and Practice". ed. 3rd. Hungary: Elsevier Science, (2007).
- Siriwardane, R.V., Shen, M.S., Fisher, E.P., and Poston, J.A., "Adsorption of CO<sub>2</sub> on Molecular Sieves and Activated Carbon". *Energy & Fuels*, Vol. 15, (2001), 279-284.
- Chue, K.T., Kim, J.N., Yoo, Y.J., Cho, S.H., and Yang, R.T., "Comparison of activated carbon and zeolite 13X for CO<sub>2</sub> recovery from flue gas by pressure swing adsorption". *Industrial Engineering Chemical Research*, Vol. 34, (1995), 591-598.
- Yong, Z., Mata, V., and Rodrigues, A.E., "Adsorption of carbon dioxide onto hydrotalcite-like compounds (HTLcs) at high temperatures". *Industrial Engineering Chemical Research*, Vol. 40, (2001), 204-209.
- Xu, R., Pang, W., Yu, J., Huo, Q., and Chen, J., "Chemistry of zeolites and Related porous materials: Synthesis and Structure". Asia: John Wiley, (2007).
- Zhang, X., Tang, D., Zhang, M., and Yang, R., "Synthesis of NaX zeolite: Influence of crystallization time, temperature and batch molar ratio SiO<sub>2</sub>/Al<sub>2</sub>O<sub>3</sub> on the particulate properties of zeolite crystals". *Powder Technology*, Vol. 235, (2013), 322-328.
- Zhang, X., Tong, D., Zhao, J., and Li, X., "Synthesis of NaX zeolite at room temperature and its characterization". Vol. 104, (2013), 80-83.
- Mintova, S., Valtchev, V., Kiricsi, I., P'al-Borb'ely, G., Nagy, J.B., and Karge, H.G., "Synthesis of Nanosized FAU-Type Zeolite Studies in Surface Science and Catalysis". Cambridge, Mass, USA: Elsevier, (1999).
- Zhan, B.Z., White, M.A., Lumsden, M., Mueller-Neuhaus, J., Robertson, K.N., Cameron, T.S., and Gharghour, M., "Control of Particle Size and Surface Properties of Crystals of NaX Zeolite". *Chemical Material*, Vol. 14, (2002), 3636-3642.
- Zhan, B.Z., White, M.A., Robertson, K.N., Cameron, T.S., and Gharghour, M., "A novel, organic-additive-free synthesis of nanometer-sized NaX crystals". *Chemical Communication*, Vol. 13, (2001), 1176-1177.
- Ma, Y., Yan, C., Alshameri, A., Qiu, X., Zhou, C., and li, D., "Synthesis and characterization of 13X zeolite from low-grade natural kaolin". *Advanced Powder Technology*, Vol. 25, (2013), 495-499.
- Ansari, M., Aroujalian, A., Raisi, A., Dabir, B., and Fathizadeh, M., "Preparation and characterization of nano-NaX zeolite by microwave assisted hydrothermal method". *Advanced Powder Technology*, Vol. 25, (2013), 722-727.
- Ngoc, D.T., Pham, T.H., and Nguyen, K.D.H., "Synthesis, characterization and application of nanozeolite NaX from

- Vietnamese kaolin". *Advances in Natural Sciences: Nano Sciences Nanotechnology*, Vol. 4, (2013), 146-157.
21. Tosheva, L. and Valtchev, V.P., "Nanozeolites: Synthesis, Crystallization Mechanism, and Applications". *Chemical Material*, Vol. 17, (2005), 2494-2513.
  22. Wang, H., Holmberg, B.A., and Yan, Y., "Synthesis of template-free zeolite nanocrystals by using in situ thermoreversible polymer hydrogels". *Journal of the American Chemical Society*, Vol. 125, (2003), 9928-9929.
  23. Karimi, R., Bayati, B., Aghdam, N.C., Ejtemaee, M., and Babaluo, A.A., "Studies of the effect of synthesis parameters on ZSM-5 nanocrystalline material during template-hydrothermal synthesis in the presence of chelating agent". *PowderTechnology*, Vol. 229, (2012), 229-236.
  24. Aly, H.M., Moustafa, M.E., and Abdelrahman, E.A., "Synthesis of mordenite zeolite in absence of organic template". *Advance Powder Technology*, Vol. 23, (2012), 757-760.
  25. Liang, Z., Marshall, M., and Chaffee, A.L., "Comparison of Cu-BTC and zeolite 13X for adsorbent based CO<sub>2</sub> separation". *Energy Procedia*, Vol. 1, (2009), 1265-1271.
  26. Cheung, O., Bacsik, Z., Liu, Q., Mace, A., and Hedin, N., "Adsorption kinetics for CO<sub>2</sub> on highly selective zeolites NaKA and nano-NaKA". *Applied Energy*, Vol. 112, (2013), 1326-1336.
  27. Fathizadeh, M., Aroujalian, A., and Raisi, A., "Effect of added NaX nano-zeolite into polyamide as a top thin layer of membrane on water flux and salt rejection in a reverse osmosis process". *Journal of Membrane Science*, Vol. 375, (2011), 88-95.
  28. Fathizadeh, M. and Aroujalian, A., "Synthesis and Characterization of Nano Particle Crystals of NaX Zeolite". *International Journal of Industrial Chemical*, Vol. 2, (2011), 140-143.
  29. Anbia, M., Hoseini, V., and Mandegarzar, S., "Synthesis and characterization of nanocomposite MCM-48-PEHA-DEA and its application as CO<sub>2</sub> adsorbent". *Korean Journal of Chemical Engineering*, Vol. 29, (2012), 1776.
  30. Anbia, M. and Mandegarzar, S., "Enhanced hydrogen sorption on modified MIL-101 with Pt/CMK-3 by hydrogen spillover effect". *Journal of Alloys and Compounds*, Vol. 532, (2012), 61-67.
  31. Salehi, S. and Anbia, M. "Investigation of Carbon Dioxide Adsorption on Amino-Functionalized Mesoporous Silica". *International Journal of Engineering*, vol. 28, (2015), 848-854.
  32. Silva, J.A.C., A.F.Cunha, K.Schumann, and Rodrigues, A.E., "Binary adsorption of CO<sub>2</sub>/CH<sub>4</sub> in binderless beads of 13X zeolite". *Microporous and Mesoporous Materials*, Vol. 187, (2014), 100-107.
  33. Kulprathipanja, S., "Zeolite In Industrial Separation and Catalysis," USA: Wiley, (2010).

## Effect of Particle Size of NaX Zeolite on Adsorption of CO<sub>2</sub>/CH<sub>4</sub>

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در کار حاضر، نانو و میکرو زئولیت NaX با استفاده از روش هیدروترمال سنتز شدند. سپس ظرفیت های جذب و ایزوترم های گازهای دی اکسیدکربن و متان در زئولیت نانو ذره سنتز شده در سه دمای ۲۸۸، ۲۹۸ و ۳۰۸ درجه کلوین و فشارهای مختلف ۱ تا ۲۰ بار تعیین می شوند. نانوزئولیت NaX ظرفیت جذب دی اکسیدکربن بالاتری نسبت به متان نشان داد. انتخاب پذیری CO<sub>2</sub>/CH<sub>4</sub> در نانوزئولیت NaX در دمای ۲۸۸ درجه کلوین و فشار ۲۰ بار، ۵/۴۷ بود. داده های آزمایشگاهی از ایزوترم لانگمویر-فرندلیچ پیروی می کنند. نتایج نشان داد که تبدیل اندازه ذره از میکرو متر به نانو متر، باعث می شود جذب دی اکسیدکربن حدود ۲۸ درصد در دمای ۲۸۸ درجه کلوین و فشار ۲۰ بار افزایش یابد ( از ۵/۰۶۷ به ۶/۵۳۶ میلی مول بر گرم).

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