

SYNTHESIS OF ZEOLITES FROM SILICA GEL WASTE FOR CARBON DIOXIDE ADSORPTION

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Abstract

Growing volumes of solid waste from industrial and agricultural sources continue to raise environmental concerns. One such waste stream is silica gel, frequently used in packaging and typically discarded, despite its high silica content. In this study, discarded silica gel was employed as a silica source for the synthesis of sodium-form zeolites, namely NaA, NaX, and NaY. The resulting materials were assessed for their performance in carbon dioxide adsorption. Crystalline zeolite phases were successfully formed, with CO₂ uptake capacities of 4.10, 5.84, and 4.39 mmol/g for NaA, NaX, and NaY, respectively. These synthesized zeolites showed adsorption properties on par with those of commercially available analogues. The findings support the transformation of silica gel waste into effective, affordable, and sustainable materials for CO₂ capture applications.

Keywords: Zeolites; Linde Type A; Faujasite; Silica gel; Carbon dioxide; CO₂

Introduction

Burning fossil fuels greatly contributes to carbon dioxide emissions, making it a primary factor in greenhouse gas buildup. To lessen this impact, capturing and utilizing CO₂ is essential for improving the environmental sustainability of fossil fuel energy systems (de Aquino *et al.*, 2020; Muriithi *et al.*, 2020). Several methods for CO₂

separation exist, including chemical absorption, physical adsorption, cryogenic distillation, membrane technologies, biological fixation, and oxy-fuel combustion with CO₂ recycling. Among these, adsorption on solid materials has become popular due to its energy efficiency, simple operation, and affordability (Kaithwas *et al.*, 2012;

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Keawkumay *et al.*, 2024). Zeolites are widely used as adsorbents because of their strong affinity for gas molecules. Their key features include a high surface area, remarkable thermal stability, low bulk density, significant pore volume, and a well-ordered crystalline framework with uniform micropores (de Carvalho Izidoro *et al.*, 2024). Structurally, zeolites are microporous aluminosilicates made of tetrahedrally coordinated TO₄ units (T = Si or Al), linked by shared oxygen atoms (Wittayakun *et al.*, 2008). Their empirical formula is $M_{a/b}[(AlO_2)_a(SiO_2)_y] \cdot cH_2O$, where M is a charge-balancing alkali or alkaline earth metal cation with valency b, c indicates water molecules, and a and y represent the number of $[SiO_4]^{4-}$ and $[AlO_4]^{5-}$ tetrahedra, respectively (Lakiss *et al.*, 2020). Zeolite pore sizes typically range from 0.3 to 1.0 nanometers. The silicon-to-aluminum ratio (Si/Al), which can be adjusted based on the framework type, strongly affects both the structural stability and adsorption performance.

A major challenge in zeolite production is the dependence on synthetic silica sources like silicon alkoxides and fumed silica, which are expensive and less environmentally friendly (Sharma *et al.*, 2015). To overcome this, researchers have investigated alternative silica-rich feedstocks from agricultural and industrial residues, such as rice husk (Wittayakun *et al.*, 2008), coal fly ash (Amoni *et al.*, 2019), rice husk ash (Kamseu *et al.*, 2017; Zhang *et al.*, 2019), and natural clays like kaolin (Tavasoli *et al.*, 2014). Silica gel waste, a by-product with high silica content, has also been recognized as a promising and underused source for zeolite synthesis (Khaosomboon *et al.*, 2018). However, comprehensive research on using silica gel waste to produce various zeolite frameworks remains limited.

Silica gel is often used in moisture control, adsorption processes, and chromatographic separations. However, improper disposal of used silica gel leads to environmental problems, especially as its use increases. Recycling this waste into useful materials offers a sustainable waste management option. Due to its high silica content, discarded silica gel is a promising renewable resource for zeolite production.

Zeolites NaA, NaX, and NaY are synthetic materials mainly distinguished by their aluminum content, which influences their framework structure and ion-exchange properties. Zeolite A, classified under the Linde Type A (LTA) structure, usually has a low Si/Al ratio between 1.0 and 1.5. Both zeolite X and Y belong to the faujasite (FAU) structural family; NaX typically has a Si/Al ratio of 1.0-1.5, while NaY has a higher ratio, above 1.5. FAU-type zeolites have pore openings around

7.4 Å, whereas LTA zeolites feature smaller openings near 4 Å. These pore sizes, together with their high surface areas, make them effective for gas adsorption. Among synthetic zeolites, LTA and FAU frameworks are commonly used for CO₂ capture due to their adjustable structures and strong adsorption capabilities.

This study aims to synthesize phase-pure NaA, NaX, and NaY zeolites using silica gel waste as the only silica source through a conventional hydrothermal method. The materials produced were analyzed for their structural, morphological, and basic properties, along with N₂ adsorption-desorption behavior. Additionally, their CO₂ adsorption performance was assessed and compared with that of commercial zeolites to understand how framework type and Si/Al ratio affect adsorption capacity.

Materials and Methods

Waste silica gel was collected from desiccant packets commonly found in furniture packaging. Elemental analysis using energy dispersive X-ray fluorescence (EDXRF, Horiba XGT-5200) showed a silica purity of 99%. The material was dissolved in a sodium hydroxide (NaOH) solution and then used as the silica precursor for zeolite synthesis. Zeolite NaA, NaX, and NaY were synthesized by modifying established procedures reported in previous studies (Jantarit *et al.*, 2020; Keawkumay *et al.*, 2019; Khaosomboon *et al.*, 2018).

Synthesis of zeolite NaA using silica gel waste

Zeolite NaA was synthesized via a hydrothermal method using a gel mixture with a molar composition of 3.165Na₂O: Al₂O₃: 1.926SiO₂: 128H₂O (Khaosomboon *et al.*, 2018). First, 12.27 g of NaOH was dissolved in 90 mL of deionized water inside a Teflon vessel. This solution was divided equally into two parts. One part was mixed with 2.04 g of silica gel waste and stirred at 90°C for 3 hours to produce a sodium silicate solution. Simultaneously, 6.36 g of anhydrous NaAlO₂ was dissolved in the remaining NaOH solution at room temperature until a clear aluminate solution was formed.

The two solutions were quickly mixed, forming a viscous gel. The mixture was sealed and stirred at room temperature for 30 minutes to achieve uniformity. The gel was then subjected to static hydrothermal crystallization at 80°C for 2.5 hours. After cooling, the product was washed multiple times with deionized water until the wash solution's pH dropped below 9. Each wash cycle

involved centrifugation at 4000 rpm for 5 minutes. The final product was dried at 90°C overnight.

Synthesis of zeolite NaX using silica gel waste

Zeolite NaX was produced through a hydrothermal process using a gel with a molar ratio of $\text{Na}_2\text{O} : 4\text{Al}_2\text{O}_3 : 16\text{SiO}_2 : 325\text{H}_2\text{O}$ (Jantarit *et al.*, 2020). To create the sodium silicate precursor, 4.83 g of NaOH was dissolved in 26.67 g of deionized water in a Teflon container, then 12.3 g of silica gel waste was gradually added. The mixture was stirred at room temperature for 18 hour.

Separately, 27.24 g of NaOH was dissolved in 264 g of deionized water, after which 4.08 g of anhydrous NaAlO_2 was added. This solution was stirred for 1 hour to ensure complete dissolution. The sodium silicate solution was then mixed with the aluminate solution and stirred for another hour to create a homogeneous solution gel.

The final gel was subjected to hydrothermal treatment at 90°C for 18 hours under static conditions. After crystallization, the solid product was washed repeatedly with deionized water until the wash solution reached a pH below 7, using multiple centrifugation steps at 4000 rpm for 5 minutes per cycle. The washed product was then dried overnight at 90°C.

Synthesis of zeolite NaY using silica gel waste

Zeolite NaY was synthesized using a hydrothermal method with a gel composition of $4.62\text{Na}_2\text{O} : \text{Al}_2\text{O}_3 : 10\text{SiO}_2 : 180\text{H}_2\text{O}$ (Keawkumay *et al.*, 2019). The sodium silicate solution was prepared by dissolving 28.7 g of silica gel waste in a mixture of 11.5 g of NaOH and 59.8 g of deionized water within a Teflon container. This mixture was stirred at room temperature for 24 hours.

To prepare the seed gel, 1.02 g of NaOH was dissolved in 5.1 g of water, followed by the addition of 0.52 g of anhydrous NaAlO_2 . After thorough mixing, 5.66 g of Na_2SiO_3 solution was added, stirred for 10 minutes, sealed, and aged at ambient temperature for 24 hours.

The feedstock gel was produced by dissolving 0.035 g of NaOH in 32.74 g of water in a separate beaker, then adding 3.27 g of NaAlO_2 . The solution was stirred until uniform before 35.61 g of Na_2SiO_3 solution was introduced and mixed for 10 minutes.

This feedstock was then combined with the aged seed gel, stirred for another 10 minutes, sealed, and allowed to age again for 24 hours at room temperature. Crystallization was performed at 90°C for 24 hours under static conditions. The resulting solid was washed repeatedly with water until the

filtrate pH dropped below 9, using centrifugation at 4000 rpm for 5 minutes per cycle. The final product was dried overnight at 90°C.

Zeolite characterization

Elemental analysis of the zeolite samples was carried out using a wavelength-dispersive X-ray fluorescence (XRF) spectrometer equipped with a Rhodium X-ray tube, operated at 50 kV and 60 mA, with a 150 μm collimator. Phase identification was determined using X-ray diffraction (XRD) on a Bruker D8 ADVANCE diffractometer with a monochromatic $\text{Cu K}\alpha$ source, operated at 40 kV and 40 mA. Data were collected at a scan rate of 0.2 s per step and a step size of 0.02° .

Surface morphology and elemental mapping were examined by scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM-EDX, JEOL JSM-6400), operated at 20 kV under a vacuum of 10^{-4} Pa with a tungsten filament. The samples were mounted on carbon adhesive tape and coated with either carbon or gold prior to imaging. Particle size distribution was estimated from SEM images using ImageJ software on 7 crystals for zeolite A, 10 particles for zeolite X and 10 particles for zeolite Y. Functional group analysis was conducted using Fourier-transform infrared spectroscopy (FTIR) in ATR mode on a Bruker Vertex 70+ RamII instrument with a resolution of 4 cm^{-1} .

Nitrogen adsorption - desorption measurements were performed using a Micromeritics ASAP 2010 instrument, within a relative pressure (P/P_0) range of 0.01 to 0.99. Before analysis, the zeolite samples were evacuated at 300°C for 8 hours under vacuum. The BET method was used to determine the total surface area, while the t-plot method calculated the micropore surface area, external surface area, and micropore volume.

For CO_2 adsorption studies, the procedure followed the method described in the literature (Keawkumay *et al.*, 2024). Zeolite samples were degassed at 300°C for 3 hours under vacuum conditions. Adsorption isotherms were measured using a BEL-CAT B chemisorption analyzer at 25 °C, with pressure ranging from 0 to 100 kPa. The volume of CO_2 adsorbed was plotted against pressure, and the adsorption capacity (in mmol/g) was calculated at 100 kPa and 25°C by converting the adsorbed gas volume into molar units.

Four adsorption isotherm models - Langmuir, Freundlich, Langmuir - Freundlich (Sips), and Toth - were employed to interpret the interaction between CO_2 molecules and the zeolite surfaces.

The Langmuir model assumes monolayer adsorption on a surface with uniform energy sites,

where each site can hold only one adsorbate molecule. Once occupied, no further adsorption can occur at that location.

$$\frac{C_e}{q_e} = \frac{1}{K_L q_m} + \frac{1}{q_m} C_e \quad (1)$$

Here, q_m (mg g⁻¹) denotes the maximum adsorption capacity, K_L (L mg⁻¹) is the Langmuir constant, and C_e is the equilibrium concentration (mg L⁻¹).

The Freundlich model describes adsorption on heterogeneous surfaces and is useful over a wide concentration range, capturing non-linear behavior.

$$\ln q_e = \frac{1}{n} \ln C_e + \ln K_f \quad (2)$$

In this equation, K_f (L mg⁻¹) and n are the Freundlich constant and heterogeneity factor, respectively.

The Toth model adjusts the Langmuir equation to more accurately describe systems with surface heterogeneity and sub-monolayer coverage. It applies to a wide range of concentrations.

$$q_e = \frac{q_m K_T C_e}{[1 + (K_T C_e)^{n_T}]^{1/n_T}} \quad (3)$$

In the equation, q_m is the maximum capacity (mmol g⁻¹), K_T is Toth constant (bar⁻¹), n_T is the heterogeneity parameter. For a perfectly homogeneous surface, $n_T = 1$.

The Sips model, or Langmuir - Freundlich model, combines the advantages of both parent models. At low concentrations, it behaves like the Freundlich model, while at high concentrations, it approximates Langmuir-type monolayer adsorption.

$$q_e = \frac{q_m (K_s C_e)^{1/n_s}}{1 + (K_s C_e)^{1/n_s}} \quad (4)$$

Here, q_m is the maximum uptake (mmol g⁻¹), K_s is the Sips constant (bar⁻¹), and n_s is the Sips exponent.

Results and Discussion

The oxide composition of the silica gel waste, determined by XRF, includes SiO₂ (95.96 wt%), with trace amounts of Na₂O (1.86 wt%), MgO (1.36 wt%), TiO₂ (0.33 wt%), CaO (0.23 wt%) and Fe₂O₃ (0.27 wt%). The high silica purity and amorphous nature of the material make it particularly suitable for zeolite synthesis, as it readily reacts with NaOH to form sodium silicate (Na₂SiO₃) (Keawkumay *et al.*, 2024). According to the previous work (Keawkumay *et al.*, 2025), silica powder contains approximately 2% moisture. However, in this study, silica pellets were used, which have significantly lower moisture content. This minimizes the potential impact of residual moisture on the synthesis process and the properties of the resulting material.

Figure 1 shows the surface morphology of both synthesized and commercial zeolites A, X, and Y. Synthesized zeolite A has a well-defined cubic shape with an average crystal size of about 0.6 ± 0.3 μm. Zeolite X appears as intergrown crystal aggregates forming compact particles around 1.7 ± 0.4 μm in size. The NaY sample shows agglomerated polyhedral structures with much smaller particles of 0.3 ± 0.05 μm. In contrast, commercial zeolite A mainly displays distinct cubic crystals of 2.1 ± 0.6 μm. Commercial samples of zeolite X and Y feature well-formed octahedral particles with consistent shapes, measuring approximately 2.2 ± 0.5 μm and 1.0 ± 0.2 μm, respectively.

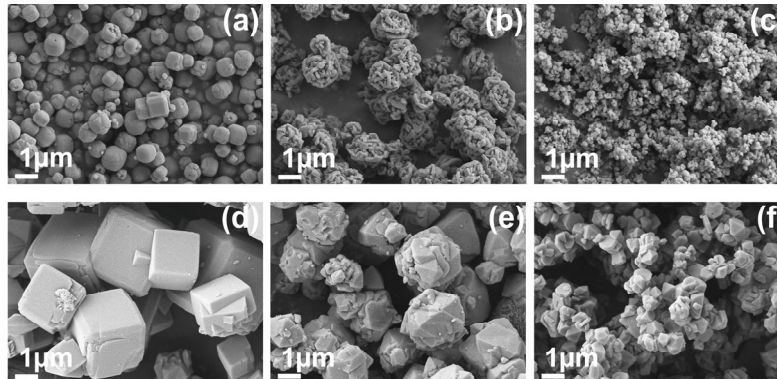


Figure 1. SEM image of synthesized zeolite A (a), synthesized zeolite X (b), synthesized zeolite Y (c), commercial A (d), commercial X (e), and commercial Y (f) by using silica gel waste as silica source with a magnification of 10 KX

Figure 2 shows the XRD patterns of the synthesized zeolite A, X, and Y, compared with their commercial counterparts. The diffraction peaks of each synthesized zeolite match well with those of the corresponding commercial standards, confirming the formation of the intended crystalline phases. In the NaX sample, minor reflections around $2\theta \approx 7^\circ$ may indicate the presence of trace impurity phases. Meanwhile, the synthesized NaY shows relatively low peak intensities, suggesting it has lower crystallinity than the commercial NaY. Overall, the XRD results confirm that silica gel waste is a viable raw material for producing phase-pure zeolite A, X, and Y.

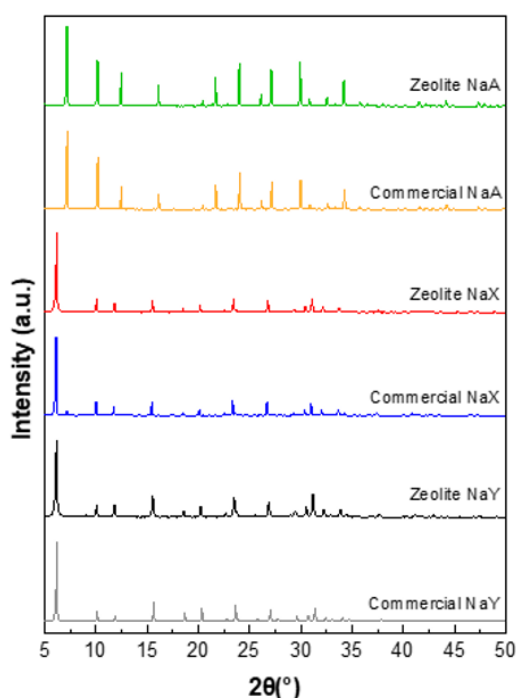


Figure 2. XRD pattern of synthesized and commercial zeolites

FTIR spectra were recorded in the range of 400-4000 cm^{-1} to analyze the vibrational characteristics of the zeolite frameworks, as shown in Figure 3. The spectra of synthesized NaA, NaX, and NaY were compared with commercial standards. Characteristic bands corresponding to T-O-T bending and symmetric stretching modes (T = Si or Al) were observed, which are only marginally influenced by the Si/Al ratio. For zeolite NaA, prominent bands were identified at 465 and 663 cm^{-1} , while those for NaX appeared at 453 and 669 cm^{-1} . The asymmetric stretching of double four-membered rings, which is more sensitive to

structural changes, appeared at 550 cm^{-1} for NaA and 561 cm^{-1} for NaX. A distinct T-O-T valence vibration at 748 cm^{-1} was evident in NaX alone. Additionally, internal asymmetric T-O stretching vibrations appeared at 987 cm^{-1} (NaA) and 976 cm^{-1} (NaX). Asymmetric T-O-T stretching (external linkages) appeared at 1096 cm^{-1} for NaA and 1082 cm^{-1} for NaX. Peaks near 1640 cm^{-1} and 3370 cm^{-1} were associated with molecular water and hydroxyl groups. These features are in line with FTIR data reported for zeolite A and X (Tsitsishvili, 2019; Keawkumay *et al.*, 2024).

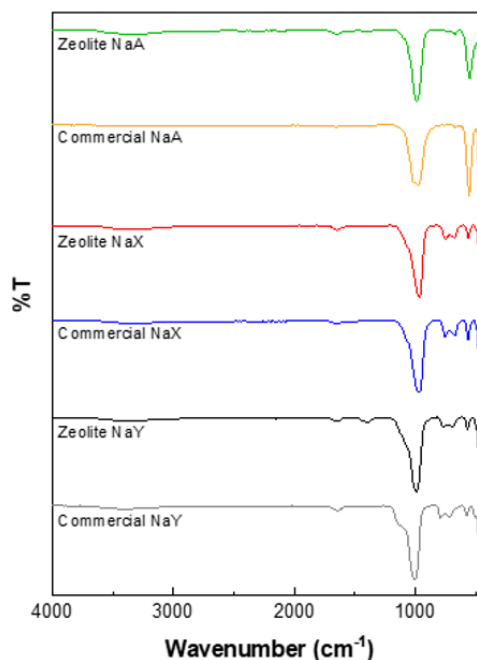


Figure 3. FTIR spectra of synthesized and commercial zeolites

Figure 4 shows the elemental mapping results from SEM-EDS analysis of the synthesized zeolites NaA, NaX, and NaY. The distribution of key elements - aluminum (Al), silicon (Si), oxygen (O), and sodium (Na) - is displayed across each zeolite particle. The mappings confirm that these elements are evenly spread within the particles, indicating consistent elemental incorporation during synthesis. The presence of Si and Al supports the formation of the aluminosilicate framework, while Na appears as the exchangeable cation balancing the negative charge of the framework. The lack of concentrated elemental clusters suggests a successful and uniform distribution of components, further confirming the structural integrity of the synthesized zeolites.

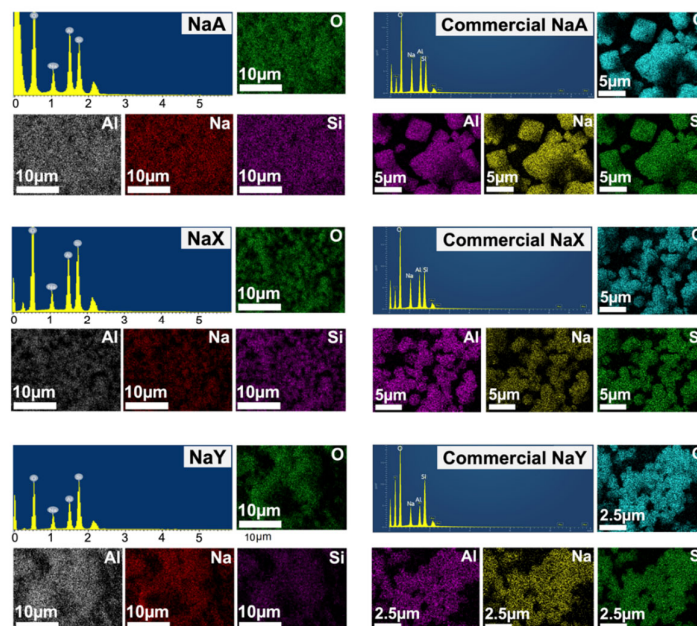


Figure 4. SEM-EDS mapping of NaA, NaX, and NaY zeolites. The synthesized zeolites display the distribution of Al, Si, O, and Na across the zeolite particles

Table 1 summarizes the silicon-to-aluminum (Si/Al) molar ratios of the synthesized zeolites, as determined by SEM-EDS and ED-XRF analyses. Zeolite A exhibited the lowest Si/Al ratio, consistent with its LTA framework, followed by zeolite X and zeolite Y, which showed progressively higher ratios. This trend is in line with their respective framework types and synthetic conditions. The Si/Al values from ED-XRF were slightly higher than those obtained from SEM-EDS, possibly due to surface sensitivity differences between the two techniques. The increase in Si/Al ratio from NaA to NaY reflects the controlled incorporation of silica into the framework during synthesis, which influences not only structural properties but also adsorption behavior in later sections.

Table 1. Si/Al ratio from ^aSEM-EDS and ^bED-XRF

Sample	Si/Al ratio ^a	Si/Al ratio ^b
Zeolite A	1.01±0.01	1.17±0.024
Zeolite X	1.46±0.033	1.67±0.023
Zeolite Y	2.11±0.155	2.25±0.007

Figure 5 presents the nitrogen adsorption-desorption isotherms for the synthesized and commercial zeolites NaX and NaY. All samples exhibit Type I(a) isotherms, which are indicative

of materials dominated by narrow micropores (pore width < 1 nm). The sharp rise in nitrogen uptake at low relative pressure (P/P_0) reflects micropore filling, a behavior characteristic of strong interactions between adsorbate molecules and the internal pore walls of microporous structures. As the pressure increases, the isotherms approach a plateau, consistent with micropore volume limitations, rather than surface area effects. Additionally, a subtle H4-type hysteresis loop appearing at $P/P_0 > 0.4$ is from aggregated crystals of zeolites. This observation aligns with the IUPAC classification for micro - mesoporous materials (Thommes *et al.*, 2015). The surface areas and micropore volumes of NaX and NaY samples are presented in Table 2.

Table 2. The surface area and pore volume of zeolites from N₂ adsorption - desorption isotherm

Sample	Surface area ^a (m ² /g)	Micropore surface area ^b (m ² /g)	Micropore volume ^b (cm ³ /g)
NaX ^s	800	744	0.27
NaX ^c	800	759	0.28
NaY ^s	669	615	0.23
NaY ^c	876	835	0.30

^aBET method,

^bt-plot method, ^ssynthesized, ^ccommercial

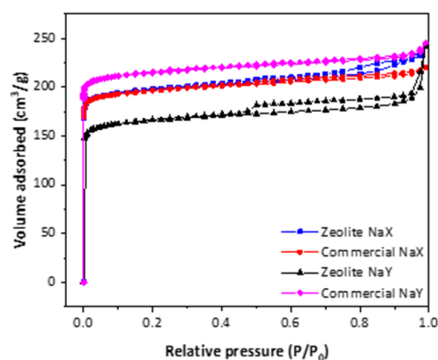


Figure 5. N₂ adsorption - desorption isotherms of zeolite X and zeolite Y using silica gel waste as silica source

The pore size profiles obtained from Non-Local Density Functional Theory (NL-DFT) analysis of N₂ sorption isotherms (Figure 6) reveal distinct textural characteristics among the zeolite samples. The synthesized NaX and commercial NaX exhibit broad mesoporous distributions with dominant pore widths centered around 4 - 6 nm. The synthesized NaY displays a sharp and intense peak around 5 nm, indicating a highly uniform mesoporous structure with concentrated pore volume. In contrast, commercial NaY presents a broader mesopore distribution similar to the NaX samples, with major contributions around 4 - 6 nm. The N₂ sorption analysis of NaA is not provided because the pore size of NaA is smaller than N₂ gas

molecules. An example of an isotherm of N₂ using zeolite LTA can be found in the literature (Keawkumay *et al.*, 2025; Obrom *et al.*, 2025).

Figure 7 presents the CO₂-TPD profiles of synthesized and commercial zeolites NaA, NaX, and NaY, revealing two main desorption regions. The first, between 150-300°C, corresponds to weak and moderate basic sites, while the second, above 300 °C, indicates the presence of stronger basic sites. The shape and intensity of these peaks reflect both the quantity and strength of surface basic sites. Table 3 summarizes the total basicity of each sample, measured in mmol CO₂ per gram. Among the materials tested, commercial NaA exhibited the highest basicity (0.139 mmol/g), followed closely by synthesized NaA (0.135 mmol/g). In contrast, both synthesized and commercial NaY showed the lowest basicity values. These trends correspond with the Si/Al ratio: lower ratios increase the number of AlO₄⁻ sites in the framework, enhancing electrostatic interactions with acidic CO₂ molecules.

Table 3. Basicity of synthesized and commercial zeolites

Sample	Basicity (mmol g ⁻¹)
Zeolite NaA	0.135
Commercial NaA	0.139
Zeolite NaX	0.052
Commercial NaX	0.068
Zeolite NaY	0.037
Commercial NaY	0.039

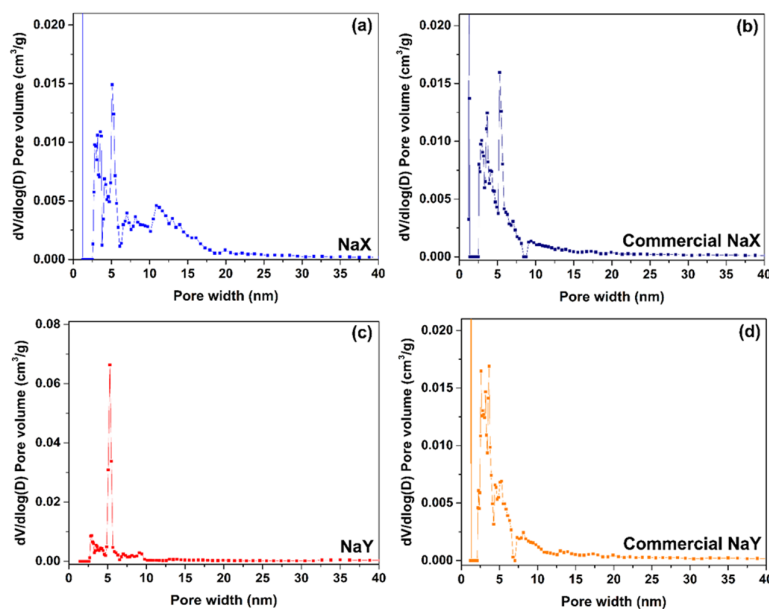


Figure 6. a.-d. the NL-DFT pore size distribution calculated from N₂ adsorption isotherms

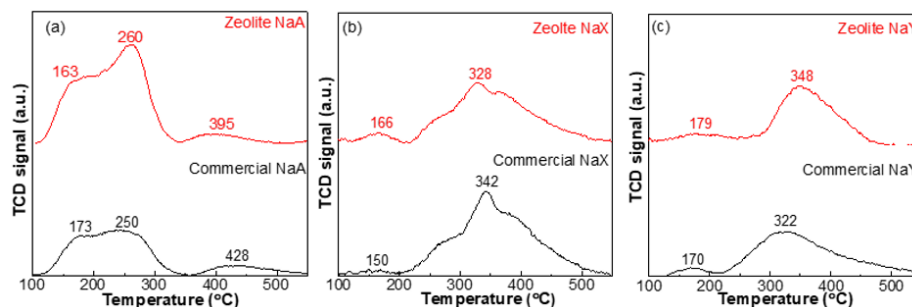


Figure 7. CO₂-TPD profiles of synthesized and commercial NaA (a), synthesized and commercial NaX (b), and synthesized and commercial NaY (c)

Figure 8 shows the CO₂ adsorption isotherms of both synthesized and commercial zeolites NaA, NaX, and NaY, measured at 25°C across a pressure range of 0 to 100 kPa. All samples exhibited Type I isotherms according to the IUPAC classification, which is typical for microporous materials that undergo physical adsorption. A rapid increase in CO₂ uptake is observed at low pressures, indicating strong interactions between CO₂ and the abundant microporous adsorption sites. As the pressure increases, the isotherms level off, suggesting that the micropores are fully occupied. The overall behavior confirms that adsorption is primarily governed by pore filling and remains stable near atmospheric pressure.

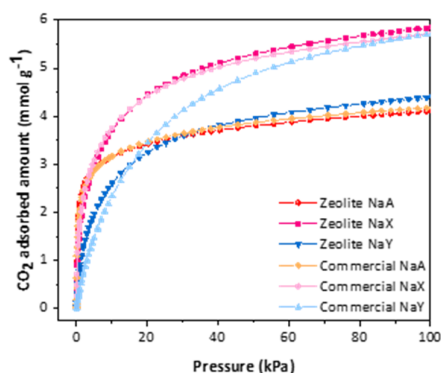


Figure 8. CO₂ adsorption isotherms of synthesized and commercial zeolites performed at 25°C

CO₂ adsorption on zeolites is primarily governed by electrostatic interactions, particularly quadrupole interactions between CO₂ molecules and the electric fields generated by the charge-balancing cations in the zeolite cavities. These cations—such as Na⁺—reside in defined sites within

the microporous channels and significantly influence adsorption strength and capacity. At low pressures, CO₂ molecules are readily attracted to these accessible cationic sites, resulting in rapid uptake. As the pressure increases and these active sites become saturated, the adsorption curve flattens, indicating micropore filling and a near-constant capacity. This behavior underscores the importance of cation location and framework structure in determining CO₂ adsorption efficiency (Siriwardane *et al.*, 2005; Bonenfant *et al.*, 2008; Keawkumay *et al.*, 2024).

As summarized in Table 4, the CO₂ adsorption capacities of the zeolites followed this trend: NaX > commercial NaX > commercial NaY > NaY > commercial NaA > NaA. This order reflects the combined effects of basicity, pore size, and framework composition on CO₂ uptake. The aluminosilicate structure of zeolites contains Lewis basic sites originating from oxygen atoms adjacent to framework aluminum. These TO₄ tetrahedra (T = Si or Al), connected through shared oxygen atoms, generate a net negative framework charge. This charge is balanced by sodium ions (Na⁺) located inside the cavities, which serve as key interaction points for CO₂ molecules. The synergistic effect of negatively charged framework sites and associated cations enhances the zeolites ability to physically adsorb CO₂ (Salehi and Anbia, 2017).

Table 4. CO₂ adsorption capacity from synthesized and commercial zeolites

Sample	CO ₂ adsorption capacity	
	(mg g ⁻¹)	(mmol g ⁻¹)
Zeolite NaA	180	4.10
Commercial NaA	183	4.17
Zeolite NaX	257	5.84
Commercial NaX	251	5.70
Zeolite NaY	193	4.39
Commercial NaY	251	5.70

Zeolites with lower Si/Al ratios contain a higher proportion of aluminum in the framework, which increases the density of charge-compensating cations and thus the number of basic sites. These sites enhance electrostatic interactions with CO₂, contributing to higher adsorption capacities and improved selectivity. As seen in Table 4, NaX - with its relatively lower Si/Al ratio compared to NaY - demonstrated superior CO₂ adsorption (5.84 vs. 4.39 mmol g⁻¹), consistent with this structural influence (Abdullahi *et al.*, 2017).

The relatively lower CO₂ uptake observed for synthesized zeolite A, compared to zeolite X, is primarily due to differences in their pore structures. Zeolite X possesses wider pore openings (~7.4 Å), which facilitate the diffusion of CO₂ molecules into its internal channels, thereby improving access to adsorption sites. In contrast, zeolite A has narrower pores (~4.1 Å), which impose greater resistance to molecular diffusion and restrict access to internal cavities. As a result, fewer CO₂ molecules are adsorbed despite the availability of active sites. Notably, the synthesized NaA samples still exhibited CO₂ adsorption capacities comparable to their commercial equivalents, as shown in Table 5.

The synthesized NaY zeolite exhibited lower CO₂ adsorption capacity than commercial NaY, primarily due to differences in crystallinity. Since CO₂ adsorption depends on crystallinity, the reduced surface area and pore volume of synthesized NaY, resulting from lower crystallinity, led to decreased adsorption (Table 3). Conversely, the synthesized NaX samples demonstrated higher adsorption capacity than their commercial

counterparts, likely due to the presence of impurity phases in commercial NaX, which negatively affect adsorption performance.

The findings clearly demonstrate that zeolites derived from silica gel waste are efficient materials for CO₂ adsorption, offering competitive performance alongside a relatively simple preparation method. Moreover, repurposing silica gel waste as a raw material presents a low-cost and sustainable strategy, with strong potential for large-scale implementation in gas separation technologies. A comparison of the adsorption capacity of CO₂ in zeolites shows in Table 5.

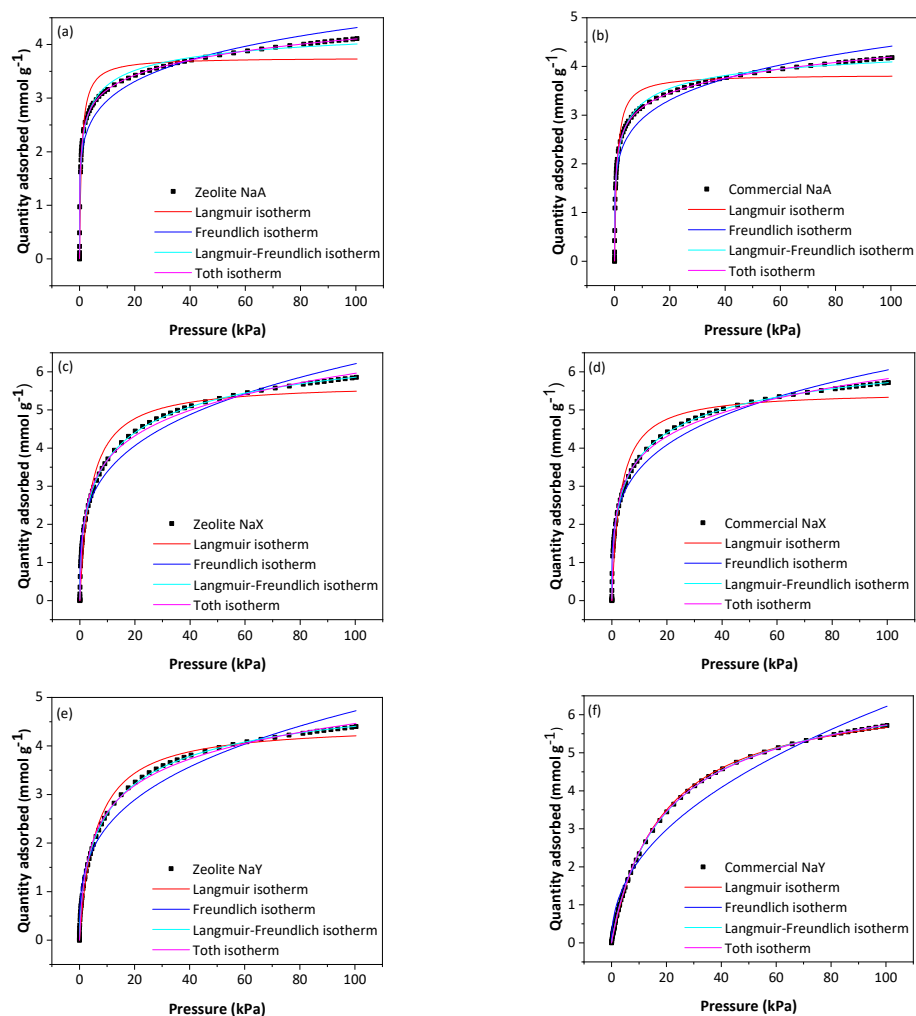
Figure 9 displays the CO₂ adsorption isotherms for both synthesized and commercial zeolites, fitted with four commonly used models: Langmuir, Freundlich, Langmuir - Freundlich (Sips), and Toth. The corresponding correlation coefficients (R²) for each model and sample are provided in Table 6. Among the models tested, the Toth model gave the best fit for NaA zeolites, with R² values exceeding 0.999 for both synthesized and commercial samples, indicating high accuracy in representing the experimental data. In contrast, the adsorption behavior of NaX and NaY - both synthesized and commercial - was best described by the Sips model, which accounts for heterogeneous surface characteristics. These findings are consistent with previous studies and confirm that the choice of isotherm model depends strongly on the zeolite structure and composition (Zouaoui *et al.*, 2021; Cui *et al.*, 2023; Keawkumay *et al.*, 2024).

Table 5. Comparison of CO₂ adsorption capacities in various zeolite types (Davaranah *et al.*, 2020).

Sample	Condition pretreatment (K)	CO ₂ adsorption capacity (mmol g ⁻¹)	Ref.
13X	593	4.6	Khraisheh <i>et al.</i> (2020)
ZSM-5	573	1.6	Li <i>et al.</i> (2013)
	373	0.9	Wang <i>et al.</i> (2017)
4A	523	1.65	Siriwardane <i>et al.</i> (2001)
NaY and KY	393	4.4-5.18	Walton <i>et al.</i> (2006)
NaA from sugarcane bagasse (SCBA)	300°C, 3 h under vacuum	1.56	Keawkumay <i>et al.</i> (2024)
NaA-P from SCBA	300°C, 3 h under vacuum	4.30	Keawkumay <i>et al.</i> (2024)
NaA-PS from SCBA	300°C, 3 h under vacuum	4.10	Keawkumay <i>et al.</i> (2024)
Commercial NaA	300°C, 3 h under vacuum	4.38	Keawkumay <i>et al.</i> (2024)
Y	No treatment	2.18	Gouveia <i>et al.</i> (2020)
4A	200°C, 12 h under vacuum	3.39	Panda <i>et al.</i> (2019)
5A	150°C, 12 h under vacuum	3.68	Gao <i>et al.</i> (2015)
NaA	300°C, 3 h under vacuum		This work
NaX	300°C, 3 h under vacuum		This work
NaY	300°C, 3 h under vacuum		This work

Table 6. CO₂ adsorption isotherm properties of commercial and synthesized zeolites

Sample	Correlation coefficients (R ²)			
	Langmuir	Freundlich	Langmuir-Freundlich	Toth
Zeolite NaA	0.95904	0.95622	0.99639	0.99910
Commercial NaA	0.96043	0.95973	0.99748	0.99952
Zeolite NaX	0.97434	0.98308	0.99984	0.99749
Commercial NaX	0.96879	0.98259	0.99982	0.99762
Zeolite NaY	0.98792	0.97886	0.99962	0.99747
Commercial NaY	0.99952	0.97746	0.99997	0.99984

**Figure 9.** CO₂ adsorption isotherms of synthesized (a) and Commercial NaA (b); synthesized (c) and Commercial NaX (d); synthesized (e) and commercial NaY (f); fitted with Langmuir, Freundlich, Langmuir-Freundlich and Toth models

Conclusions

This study successfully demonstrates the synthesis of phase-pure zeolites A, X, and Y in sodium form from silica gel waste using a conventional

hydrothermal method. The resulting materials exhibited competitive CO₂ adsorption capacities - 4.10 mmol/g for NaA, 5.84 mmol/g for NaX, and

4.39 mmol/g for NaY - with synthesized NaX notably outperforming its commercial counterpart. Adsorption isotherm fitting indicated that the Toth model best described NaA adsorption behavior, while the Sips model provided the most accurate fit for NaX and NaY. These findings highlight the potential of silica gel waste as a sustainable and low-cost silica source for producing efficient CO₂ adsorbents, offering both environmental and economic benefits for scalable gas separation applications.

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