

Gas Separation

Inverse Adsorption Separation of N₂O/CO₂ in AgZK-5 Zeolite

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Abstract: Nitrous oxide (N₂O), as the third largest greenhouse gas in the world, also has great applications in daily life and industrial production, like anesthetic, foaming agent, combustion supporting agent, N or O atomic donor. The capture of N₂O in adipic acid tail gas is of great significance but remains challenging due to the similarity with CO₂ in molecular size and physical properties. Herein, the influence of cation types on CO₂-N₂O separation in zeolite was studied comprehensively. In particular, the inverse adsorption of CO₂-N₂O was achieved by AgZK-5, which preferentially adsorbs N₂O over CO₂, making it capable of trapping N₂O from an N₂O/CO₂ mixture. AgZK-5 shows a recorded N₂O/CO₂ selectivity of 2.2, and the breakthrough experiment indicates excellent performance for N₂O/CO₂ separation. The density functional theory (DFT) calculation shows that Ag⁺ has stronger adsorption energy with N₂O, and the kinetics of N₂O is slightly faster than that of CO₂ on AgZK-5.

As the third largest greenhouse gas after carbon dioxide (CO₂) and methane (CH₄), nitrous oxide (N₂O) with a global warming potential (GWP) of about 300 causes a 300 times higher greenhouse effect than CO₂ and the concentration in the atmosphere has reached 333.2 ± 0.1 ppb in 2020.^[1] It is worth noting that N₂O is also a valuable chemical which can be used as anesthetic, foaming agent, combustion supporting agent, N or O atomic donor, etc.^[2] About 40% of total N₂O emissions come from human activities, which arise from agriculture, transportation and industrial activities.^[3] Industrially, N₂O is emitted as a by-product in the production of adipic acid. At present, thermal decomposition, catalytic decomposition and oxidation of benzene to phenol were used for the removal of N₂O.^[4]

However, these processes not only cause secondary pollution and high energy consumption but also waste the N₂O resources. Therefore, finding a cost-effective method to separate or enrich N₂O for other industrial applications is urgent and meaningful.

Particularly worth mentioning is that in addition to N₂O whose mass fraction is 38.4 wt % in adipic acid tail gas, CO₂ (6.1 wt %), N₂ (47.7 wt %), O₂ (5.6 wt %), H₂O (1.7 wt %), CO (0.3 wt %) and NO₂ (0.2 wt %) also exist in the tail gas, which shows that it is a big challenge to capture N₂O from this mixed tail gas.^[5] Pressure swing adsorption (PSA) technology based on porous adsorbents catches a lot of attention in recent years because of its simple operation and low energy consumption.^[6] For most adsorbents, the adsorption uptake order is H₂O or NO₂ > CO₂ or N₂O > CO > N₂ or O₂.^[7] The mass contents of H₂O, CO and NO₂ are trace amounts and the physical properties are quite different from N₂O, so they are easy to remove upfront. Therefore, the CO₂/N₂O/N₂/O₂ mixture separation is the most important problem to be solved.

The physical property difference of N₂O towards N₂ and O₂ is obvious, so it is easy to separate.^[7c,8] However, the molecular weight of CO₂ is 44 g/mol which is the same as the isoelectronic N₂O, also they are both linear molecules with a dynamic diameter of 3.3 Å. In addition, the boiling point (CO₂: 194.7 K; N₂O: 184.7 K) and polarizability (CO₂: 29.1 × 10²⁵/cm³; N₂O: 30.3 × 10²⁵/cm³) are very similar.^[8b] In conclusion, the CO₂-N₂O separation is a great challenge for the selection of adsorbent. If CO₂ is removed first, two steps are needed to separate N₂O: CO₂/N₂O and then the N₂O/N₂ or O₂ separation. Obviously, the most important challenge is the mixture of CO₂/N₂O. So far, CO₂/N₂O separation can be effectively achieved by modifying MIL-100Cr with ethylenediamine and by regulating cations in zeolites.^[9] However, if N₂O is directly preferentially adsorbed, high purity N₂O can be obtained by just one step. Thus, the most important aspect is the separation of N₂O/CO₂, which is simpler and more energy-saving, but the requirements for adsorbents are more stringent. For the separation of N₂O/CO₂, some flexible metal-organic frameworks (MOFs) can achieve preferential adsorption of N₂O, but the separation is not very efficient.^[5,10] MIL-101Cr-NH₂ and MIL-100Fe with exposed Fe²⁺ unsaturated metal sites after high temperature and high vacuum activation can achieve preferential adsorption of N₂O and effective separation of N₂O/CO₂, but the selectivity is lower than 2.^[7e,f] To this end, it is still very important to find an adsorbent with higher selectivity for N₂O.

As an important means to change the character of zeolite, ion exchange has a great influence on the adsorption and separation properties.^[7d,11] Ag⁺ exchanged zeolites and

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loaded MOFs materials have been researched widely for the separation of olefin from paraffin as Ag^+ is capable of π -complexation with olefins.^[12] Also, Ag^+ exchanged zeolites can achieve efficient hydrogen isotope separation and N_2/CH_4 separation.^[13] In the hard-soft-acid-base concept, CO_2 and N_2O possess quite similar moderate chemical hardness as Lewis bases (8.8 and 7.6 eV, respectively).^[14] Ag^+ is a soft Lewis acid with chemical hardness of 6.96 eV and N_2O is a slightly softer Lewis base as compared to CO_2 , so N_2O may have a stronger interaction with Ag^+ .

In this work, the adsorption and separation performance of CO_2 and N_2O on Ag^+ exchanged 8-membered ring KFI-type zeolite ZK-5 was studied. Compared with the original zeolite KZK-5, the modified zeolite AgZK-5 achieved adsorption reversal, from CO_2 -selective adsorbent to N_2O -selective adsorbent.

The KFI structure contains 8-hedral double 6-rings (d6rs), 18-hedral pau cages, and 26-hedral lta cages, wherein the window size of its 8-ring is $3.9 \times 3.9 \text{ \AA}$ which is suitable for N_2O and CO_2 molecular adsorption (Figure S1).^[15] Powder X-ray diffraction (PXRD) confirmed that the structures of AgZK-5 are maintained well compared with the original zeolites (Figures S2–3). The content and homogeneous distribution of silver were confirmed by inductively coupled plasma-optical emission spectroscopy (ICP-OES, Tables S1–3) and energy dispersive spectrum (EDS, Figures S4–6). The water molecules in the pore of AgZK-5 could be removed completely by heating to 523 K (Figures S7–8). The N_2 adsorption capacity at 77 K and Brunauer–Emmett–Teller (BET) surface area of ZK-5 zeolite changes from $85 \text{ cm}^3/\text{g}$ to $123 \text{ cm}^3/\text{g}$ and $298 \text{ m}^2/\text{g}$ to $434 \text{ m}^2/\text{g}$ respectively after K^+ is exchanged with Ag^+ (Table S4 and Figures S9–11), due to the smaller size of Ag^+ than K^+ (radius Ag^+ : 1.15 \AA , K^+ : 1.38 \AA).

The state of silver was confirmed by X-ray photoelectron spectroscopy (XPS) and ultraviolet and visible spectrophotometry (UV/Vis), and all of them indicate that silver exists mainly in the state of monovalent silver in the zeolites (Figures S12–15).^[16] However, those methods, particularly XPS, detect silver only on in the surface layer. In order to assess the average localization of silver thorough the sample, Rietveld refinement of powder X-ray diffraction data was attempted. A preliminary check confirmed the data consistency via Le Bail fitting (Figure S16), which showed that the AgZK-5 is nearly phase-pure and retains almost precisely the unit cell parameters ($I\bar{m}\bar{3}m$, 18.686 \AA vs 18.578 \AA for ZK-5). The Rietveld refinement (Figure S17, Bragg $R = 11.7\%$) suggests seven Ag locations (Tables S5–6), with three of them representing the major silver content (Figures S18d and S19–25): Ag1, somewhat out of the 8-membered ring plane towards the center the spherical pore (Figure S18a); Ag2, slightly offset from the planes of the double 6-membered ring, and delocalized over three close positions related by a 3 axis (Figure S18b); and Ag3, close to the interior of the spherical cavity (Figure S18c). It is important to note that the Rietveld refinement gives only a quite approximate silver atoms positions, which is reflected by the high refined thermal displacement factor (in other words, the most probable localizations of the Ag atoms are not

confined to small areas, but covers significant volumes). As for the nature of those silver atoms, Ag2 unambiguously represents Ag^+ ions, Ag3 represents small silver clusters randomly scattered in the pores, and Ag1 could represent both. The primary location of confined K^+ and Ag^+ sites in ZK-5 zeolite were also studied by density functional theory (DFT) calculations, as shown in Figures 1 and S26. Both K^+ and Ag^+ have the lowest energy out of the 8-membered ring plane of ZK-5, which is 0.00 eV , indicating that K^+ and Ag^+ are most stable in this location. However, the Rietveld refinement strongly suggests that there are delocalized silver clusters closer to the center of the pores. The DFT calculations not explicitly accounts for those clusters, but focuses on the localization of the single-atom Ag sites, which are assumed to be the catalytically most productive ones (corresponds primarily to Ag2 in the refined structure).

To evaluate the separation potential of Ag^+ exchanged zeolites for CO_2 and N_2O , single-component adsorption isotherms for CO_2 , N_2O and N_2 on parent and Ag^+ exchanged zeolites were collected at 298 K and 1 bar (Figures 2a–c and S27–29). Clearly, before the Ag^+ exchange, CO_2 adsorption capacities on KZK-5, NaX and KChabazite exceed the capacity of N_2O , indicating a CO_2 -selective property and the $\text{CO}_2/\text{N}_2\text{O}$ selectivity at 298 K and 1 bar are calculated by ideal adsorbed solution theory (IAST) after fitted by the dual-site Langmuir equation, which are 2.2, 2.1 and 3.0, respectively (Figure 2d, S37 and Table S7). The fitting curves and parameters are shown in Figures S30–32, S36 and Tables S8 and S11. Interestingly, after the Ag^+ exchange, the adsorption capacity of CO_2 and N_2O was inverted on AgZK-5, AgX and AgChabazite. Specifically, the adsorption capacity of N_2O became higher than that of CO_2 in Ag-zeolites. Thus, after the Ag^+ exchange the modified zeolites AgZK-5, AgX and AgChabazite represent N_2O -selective adsorbents and their $\text{N}_2\text{O}/$

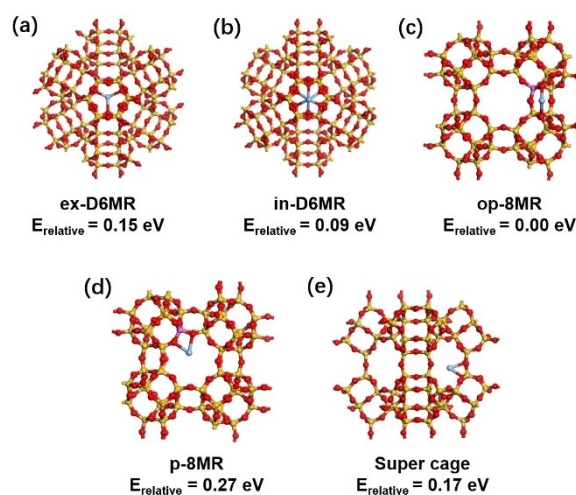


Figure 1. Relative energies of Ag^+ in different positions of ZK-5 zeolite: (a) Ag^+ in the exterior of the double 6-membered ring, (b) Ag^+ in the interior of the double 6-membered ring, (c) Ag^+ out of the 8-membered ring plane, (d) Ag^+ in the 8-membered ring plane and (e) Ag^+ in the super cage. (O: red, Si: yellow, Al: pink and Ag: light blue)

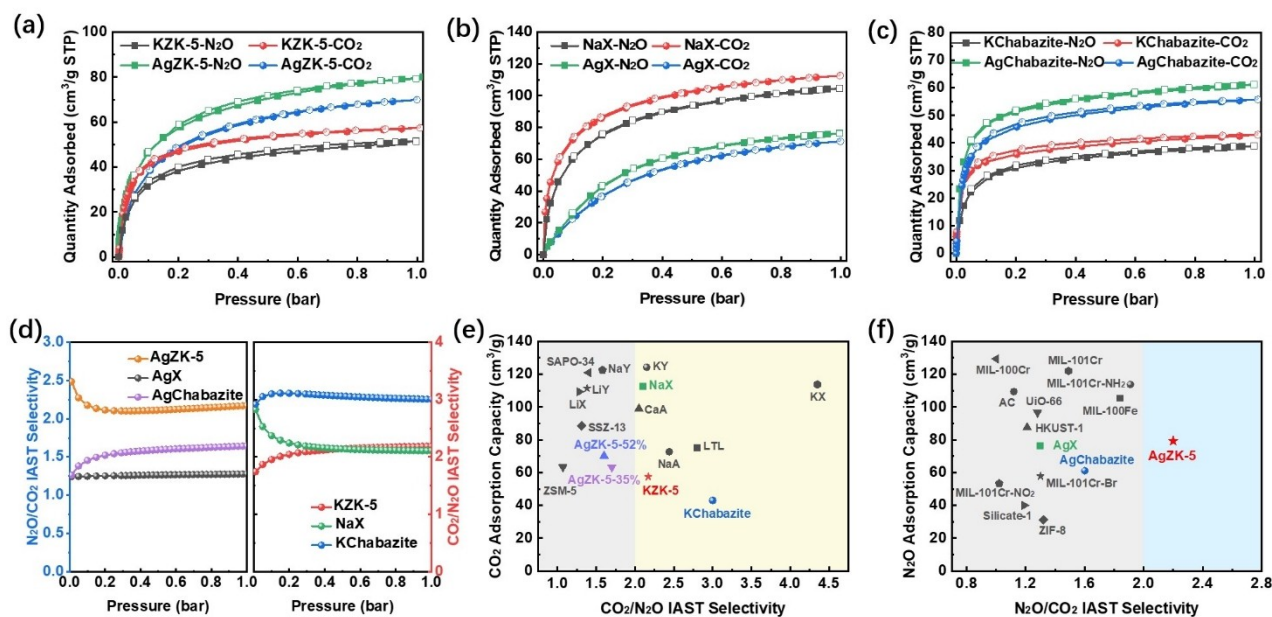


Figure 2. CO₂ and N₂O sorption isotherms at 298 K on (a) KZK-5 and AgZK-5, (b) NaX and AgX, (c) KChabazite and AgChabazite (filled symbols adsorption, empty symbols desorption), (d) IAST selectivity of CO₂/N₂O or N₂O/CO₂ on different zeolite with different cations, (e) CO₂/N₂O adsorption selectivity plotted against CO₂ uptake at 298 K and 1 bar for some benchmark adsorbents and (f) N₂O/CO₂ adsorption selectivity plotted against N₂O uptake at 298 K and 1 bar for some benchmark adsorbents.

CO₂ selectivity at 298 K and 1 bar are 2.2, 1.2 and 1.6, respectively. In addition, N₂O can be good desorption, indicating no decomposition of N₂O upon the adsorption process. It is worth noting that when the degree of Ag⁺ exchange in ZK-5 is 35% and 52% (AgZK-5-35% and AgZK-5-52%), the modified ZK-5 zeolites are still CO₂-selective adsorbents (Figure S33), but CO₂/N₂O selectivity decreases from 2.2 to 1.6 (Figure S34). This shows that the adsorption reversal of CO₂-N₂O can be realized only when the exchange degree reaches a certain level. ZK-5 with different cations have different adsorption separation properties. When the cation is K⁺, KZK-5 shows preferable CO₂/N₂O selectivity and when the cation is Ag⁺, AgZK-5 exhibits high N₂O/CO₂ selectivity (record data) as shown in Figures 2e–f and Tables S9–10, indicating an excellent adsorbent for the separation of CO₂-N₂O binary mixture, especially the separation of N₂O/CO₂. The adsorption heats of N₂O and CO₂ on KZK-5 and AgZK-5 were calculated by the virial equation using the 298 K, 308 K and 318 K adsorption isotherms (Figures S35 and S38–39), which changes from $Q_{st(\text{CO}_2)} > Q_{st(\text{N}_2\text{O})}$ on original zeolite to $Q_{st(\text{N}_2\text{O})} > Q_{st(\text{CO}_2)}$ on Ag⁺ exchanged zeolite (Figure S40). The desorption peak of N₂O on temperature-programmed desorption (TPD) was observed in the temperature range of 40–200 °C, indicating the complete regeneration of these zeolites upon thermal treatment at 200 °C or higher (Figures S41–43). In situ Fourier transform infrared spectra (In situ FTIR) with the gradual increase of N₂O flow indicates N₂O is successfully adsorbed on AgZK-5, and the adsorbed N₂O increases gradually with the increase of flow rate (Figures S44–46).

To understand the above reverse adsorption phenomenon in more detail, the adsorption energies of CO₂ and N₂O with K⁺ and Ag⁺ located out of the 8-membered ring plane (Ag1) and exterior of the double 6-membered ring (Ag2) were calculated (Figures 3 and S47–48). The adsorption energies of K⁺ with CO₂ and N₂O (N end) on Ag1 are −0.47 eV and −0.48 eV, showing that K⁺ has a similar

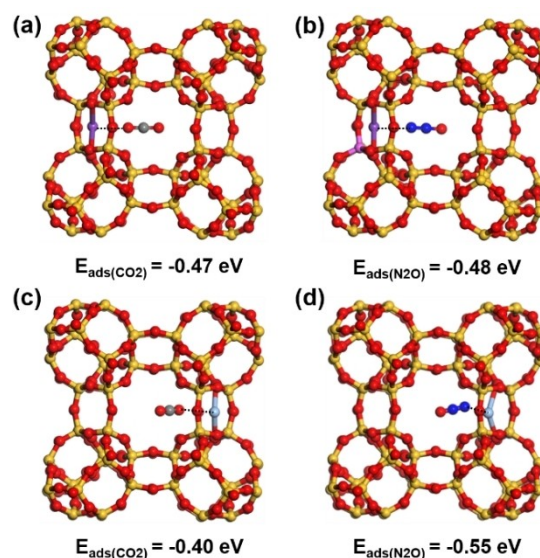


Figure 3. Adsorption energies of CO₂ and N₂O on ZK-5 with different cations: (a) K⁺-CO₂, (b) K⁺-N₂O, (c) Ag⁺-CO₂ and (d) Ag⁺-N₂O. (O: red, Si: yellow, C: grey, N: dark blue, Ag: light blue, K: purple)

interaction force with CO₂ and N₂O (Figures 3a and 3b). However, when the original cation (K⁺/Na⁺) was exchanged by Ag⁺, the adsorption energy shows a remarkable difference. The adsorption energy of Ag⁺ with CO₂ and N₂O (N end) are −0.40 eV and −0.55 eV, respectively (Figures 3c and 3d), indicating that Ag⁺ has a stronger interaction with N₂O than CO₂, which is in accordant with the above mentioned N₂O-selective adsorption material after Ag⁺ exchange. In order to further verify that Ag⁺ interacts with N₂O through N end, in situ FTIR of N₂O and Ag⁺–N–N–O or K⁺–N–N–O in ZK-5 were simulated (Figure S49). Results show that when N₂O is adsorbed on the zeolites, the wavelength of N₂O shifts towards to the long wavelength, from 2347 cm^{−1} of N₂O to 2387 cm^{−1} of Ag⁺–N–N–O in ZK-5, which is consistent with the experiment result, proving N₂O interacts with Ag⁺ through the N end.^[17]

The gas diffusion kinetic data of ZK-5 with different cations were collected on an intelligent gravimetric analyzer (Hiden IGA 001). For KZK-5, CO₂ reaches equilibrium slightly faster than N₂O (CO₂: ca. 23 min; N₂O: ca. 42 min), which explains the reason for CO₂-selective adsorption on KZK-5. (Figure 4a). Notably, the time to reach the equilibrium of N₂O on AgZK-5 is shorter than for CO₂ (N₂O: ca. 8 min; CO₂: ca. 10.5 min), which is consistent with the adsorption data and DFT calculated result.

These excellent results for equilibrium and kinetic adsorption encouraged us to further evaluate the performance of AgZK-5 in the N₂O/CO₂ separation process. The samples were pressed and granulated, with almost no adsorption loss (Figures S50–51). Breakthrough experiments were carried out using a fixed bed packed with different zeolites pellets at 298 K and a total inlet pressure of 1 bar

with a N₂O/CO₂ mixture (50%/50%, v/v) as the feed gas. The original zeolites KZK-5, NaX and KChabazite are all excellent CO₂ selective adsorbents with N₂O showing preferential breakthrough through the adsorption column (Figures 4b and S52–54). After Ag⁺ exchange, N₂O is preferentially adsorbed on Ag-zeolites and the CO₂ breakthrough time is earlier than N₂O (Figures 4c and S55–56). The breakthrough curves of AgZK-5 with different flow are shown in Figure 4c. The results shows that the retention time (retention time = N₂O breakthrough time minus CO₂ breakthrough time) and R (R = retention time/CO₂ breakthrough time) increase with the increase of the gas flow rate from 8 mL/min to 15 mL/min (Figure 4d). When the gas flow rate is 15 mL/min, the retention time and R reach the maximum, which are 3 min and 0.11 respectively, indicating that AgZK-5 has the best separation performance at 15 mL/min. When the gas flow rate reaches 20 mL/min, the retention time and R decrease sharply. Finally, the adsorption and desorption cycle experiments, PXRD after pelleting and adsorption and the breakthrough experiment of another batch AgZK-5 show that it has a good stability which can meet the demand in a real process (Figures 4e and S57–58).

In summary, an unprecedented N₂O/CO₂ separation strategy has been developed, based on Ag⁺ exchanged zeolites. Efficient separation of N₂O/CO₂ was achieved due to the stronger interaction of N₂O with the Ag⁺ cation within the zeolite micropores and the dynamics of N₂O is faster than CO₂ on AgZK-5. To the best of our knowledge, the obtained AgZK-5 exhibits the highest N₂O/CO₂ selectivity hitherto recorded among adsorbents. Breakthrough experiments demonstrated that N₂O could be efficiently separated from a N₂O/CO₂ (50:50, v/v) mixture at a flow of

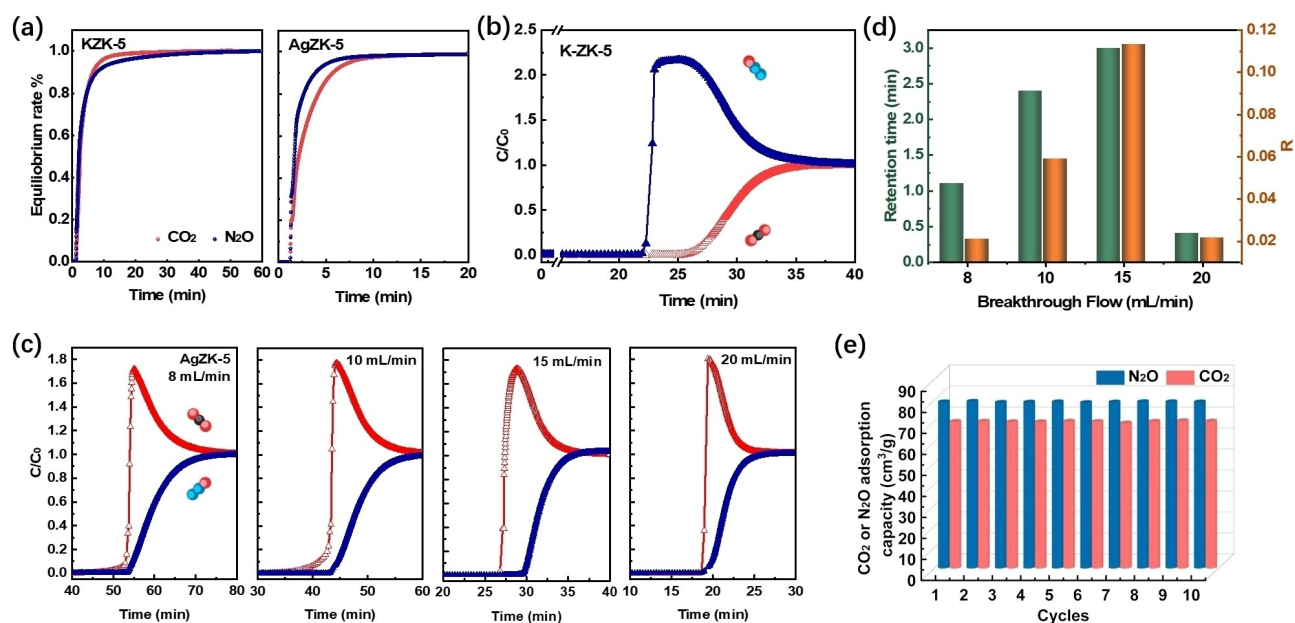


Figure 4. (a) Kinetic curves of CO₂ and N₂O on KZK-5 and AgZK-5. (b) Breakthrough curve of a CO₂/N₂O mixture (50%/50%) on KZK-5 at 15 mL/min. (c) Breakthrough curves of a N₂O/CO₂ mixture (50%/50%) with different flow on AgZK-5. (d) Comparison of retention time and R (R = retention time/CO₂ breakthrough time) on AgZK-5 with different breakthrough flow and (e) comparison of cyclic N₂O and CO₂ adsorption capacity on AgZK-5.

15 mL/min. The realization of this strategy provides a high-performance adsorbent for industrial N₂O enrichment.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

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