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Dynamic separation of CO2 from N2 using alkali-metal forms of nanosized chabazite

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Due to the rising atmospheric concentration of CO2 from human activities, the separation of CO2 from N2, commonly referred to as flue gas, has become a crucial priority.[1] There are four prevalent technologies used for CO2 capture: (i) adsorption with amine-based solvents, (ii) adsorption by nanoporous solids, (iii) cryogenic distillation, and (iv) membrane separation. Zeolites, among the materials considered for CO2 adsorption, offer the advantage of being inorganic, non-toxic substances with high thermal stability and selectivity, which can be adjusted by their framework structure and chemical composition.[1] Moreover, recent findings indicate that zeolites exhibit flexible structures.[2] This flexibility in zeolites is observable as a response to the adsorption or desorption of guest molecules. It can manifest as changes in the zeolite lattice parameters (framework dynamics) or by the relocation of extra-framework cations within zeolite pores (extra-framework dynamics).[1,2] Traditional zeolites face diffusion limitations of guest molecules through their pore networks due to their typical existence as micron-sized polycrystalline powders.[3] To overcome these limitations, various methods have been developed to increase the surface area/volume ratio. Among these approaches, nanozeolites consisting of discrete nanoparticles that result in a greater external surface area and a higher number of available active sites.[3]

We have successfully demonstrated the outstanding CO2 capture capabilities of nanosized chabazite (CHA) zeolites in various alkali forms (Na+, K+, and Cs+).[1,3,4] In this study, we initially estimated CO2 and N2 equilibrium adsorption isotherms through Grand Canonical Monte Carlo (GCMC) calculations at 298 K. Subsequently, utilizing molecular dynamics simulations, we determined the self-diffusivities of CO2 molecules at different loadings for various CHA nanocrystals. The experimental validation of dynamic CO2/N2 separation was conducted through breakthrough measurements, simulating a 17/83 (CO2/N2) mixed-component gas mixture package at 298 K (molar basis).

Based on the breakthrough results, we obtained dynamic saturation CO2 loadings of 2.48, 1.72, and 0.57 mmol g-1 for Na-CHA, K-CHA, and Cs-CHA nanosized zeolites, respectively, with CO2/N2 molar selectivity at saturation of 62, 46, and 23. Comparing the nanosized (60 nm) Cs-CHA zeolite with its micron-sized (3 μ m) counterpart, we observed significantly faster CO2 breakthrough kinetics for the nanosized Cs-CHA zeolite. Ultimately, this accelerated kinetic behavior led to a remarkable over 150% improvement in dynamic CO2 removal.

In summary, different alkali forms of nanosized CHA zeolites prove to be exceptional materials for effectively separating CO2 from N2.

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References

[1] S. Ghojavand, B. Coasne, E.B. Clatworthy, R. Guillet-Nicolas, P. Bazin, M. Desmurs, L. Jacobo Aguilera, V. Ruaux, S. Mintova, Alkali Metal Cations Influence the CO2 Adsorption Capacity of Nanosized Chabazite: Modeling vs Experiment, ACS Appl. Nano Mater. 5 (2022) 5578–5588. https://doi.org/10.1021/acsanm.2c00537.
[2] S. Ghojavand, E. Dib, S. Mintova, Flexibility in zeolites: origin, limits, and evaluation, Chem. Sci. (2023). https://doi.org/10.1039/D3SC03934J. [3] S. Ghojavand, E.B. Clatworthy, A. Vicente, E. Dib, V. Ruaux, M. Debost, J. El Fallah, S. Mintova, The role of mixed alkali metal cations on the formation of nanosized CHA zeolite from colloidal precursor suspension, J. Colloid Interface Sci. 604 (2021) 350–357. https://doi.org/10.1016/j.jcis.2021.06.176.
[4] S. Ghojavand, E. Dib, J. Rey, A. Daouli, E.B. Clatworthy, P. Bazin, V. Ruaux, M. Badawi, S. Mintova, Interplay between alkali-metal cations and silanol sites in nanosized CHA zeolite and implications for CO2 adsorption, Commun. Chem. 6 (2023) 1–8. https://doi.org/10.1038/s42004-023-00918-1.

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