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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 µm) 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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Primary authors: Dr GHOJAVAND, Sajjad (LCS, ENSICAEN); Dr MINTOVA, Svetlana (LCS, CNRS)

Co-authors: Dr COASNE, Benoit (LiPhy); Dr CLATWORTHY, Edwin (LCS); Dr KUMAR-GANDHI, Parveen (TotalEnergies); Dr GUILLET-NICOLAS, Rémy (LCS); Dr PUGNET, Veronique (TotalEnergies)

Presenter: Dr GHOJAVAND, Sajjad (LCS, ENSICAEN)

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