Advanced design of zeolitic materials for the adsorption/separation of CO_2, N_2, and CH_4 molecules

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Abstract
This thesis is dedicated to optimizing small-pore zeolite materials for efficient CO2 adsorption and gas separation. The work focuses on improving zeolite performance through cation exchange, crystal size control, and the synthesis of novel adsorbents. Mg-exchanged Gismondine (GIS) zeolites were synthesized, demonstrating enhanced CO2/N2 and CO2/CH4 selectivities due to partial cation exchange with Mg2+, which influenced the framework distortion and improved both adsorption capacity and selectivity, underscoring their potential for practical carbon capture applications. A seed-assisted synthesis method was also employed to develop nanosized zeolite P (GIS), which significantly improved CO2 adsorption kinetics by reducing diffusion limitations, with faster diffusion resulting from an intracrystalline diffusion mechanism. This superior diffusion within the nanosized zeolites led to enhanced performance in dynamic gas separation compared to their micron-sized counterparts. The thesis also introduces Levyne (LEV) zeolites as novel candidates for CO2 adsorption, where careful control of the Si/Al ratio and the use of nanosized seeds allowed for precise tuning of adsorption properties. The study highlights the critical role of the Si/Al ratio in optimizing CO2 selectivity, uptake, and adsorption−desorption behaviour, establishing LEV as a promising material for gas separation. These findings demonstrate how tailored synthesis methods, cation exchange optimization, crystal size control, and Si/Al ratio adjustment can significantly enhance the performance of zeolite-based materials in carbon capture and energy-efficient gas separation technologies.