**Revisit the Molecular Sieving Mechanism in LTA Zeolites: Does Size Really Matter?**

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“Molecular sieving”-based separation of similar-sized gases (e.g., CO2, N2, and CH4) is desirable but challenging as the “sieve” (adsorbent) with the right pore size that readily allows for exclusive admission is difficult to obtain. The “molecular trapdoor effect” can help solve this challenge, instead of relying on size-sieving, by realizing exclusive admission based on the difference in the ability of gases to dynamically open the “door” by interacting with the “door-keeper” of adsorbents. Here we show the potassium form zeolite LTA with a Si/Al ratio of 2.2 (referred to as r2KLTA) can exhibit the molecular trapdoor mechanism, as demonstrated for CO2/N2 separation and validated by gas adsorption and Powder X-ray diffraction experiments. Such an exciting separation is enabled by a K+, serving as the door-keeper, located at the eight-membered ring (8MR) pore aperture of LTA. This work affords new knowledge in understanding the mechanism of the classical zeolite molecular sieve (3A) for gas separation, that the exclusive admission of gas molecules is regulated by a dynamic door-opening rather than the static sieving. Meanwhile, the molecular trapdoor effect demonstrated on r2LTA zeolites also offers a great potential in developing adsorbents with high selectivity and elevated kinetics at proper working temperature.