Epoxide-Functionalization of Polyethyleneimine for Scalable Synthesis of Extra-Stable CO_2 Adsorbent in Temperature Swing Adsorption

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Amine-containing adsorbents have been extensively investigated for post-combustion CO_2 capture due to their ability to chemisorb low-concentration CO_2 from a wet flue gas. However, earlier studies have focused primarily on the CO_2 uptake of adsorbents, and have not demonstrated effective adsorbent regeneration and long-term material stability under such conditions.

Here we report the highly versatile and scalable synthesis of a functionalized-polyethyleneimine/silica adsorbent which simultaneously exhibits a large CO_2 working capacity (2.2 mmol/g) and long-term stability in a practical temperature swing adsorption (TSA) process, enabling the separation of concentrated CO_2. We demonstrate that the functionalization of polyethyleneimine (PEI) with 1,2-epoxybutane reduces the heat of CO_2 adsorption and facilitates CO_2 desorption (>99%) during regeneration compared to unmodified PEI (76%). Most notably, the functionalization dramatically improves long-term adsorbent stability over repeated TSA cycles due to the great suppression of urea formation and oxidative amine degradation.