Title: Synthesis of ethylene urea using carbon-dioxide-adsorbed titanium–zirconium mixed oxides

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Abstract: To address the increasing global levels of CO_2 emissions, it is necessary to develop efficient strategies that can convert CO_2 into useful chemicals such as carbonates, ureas, and carbamates. Hybrid systems that combine direct capture and subsequent utilization of CO_2 offer a unique emission-controlling pathway for achieving a carbon-neutral society. Ti-Zr mixed oxides could be ideal candidates for CO_2 adsorption owing to the high specific surface areas pore sizes. Therefore, in this study, metal oxides with desirable acid-base sites that is, $Ti_xZr_{(1-x)}O_2$ with different Ti/Zr ratios were synthesized using combined sol-gel and solvothermal methods and then utilized to adsorb and synthesize ethylene urea (EU), an important precursor of pharmaceuticals and agricultural products. The results indicated that the CO_2 adsorption capacity of TiO_2-ZrO_2 mixed oxides were higher than that of TiO_2 alone at 100 kPa and 25 °C, corresponding to their higher specific surface areas and pore volumes compared to their individual oxide counterparts. CO_2-adsorbed $Ti_xZr_{(1-x)}O_2$ was subsequently used as a CO_2 source to generate EU through a reaction with ethylenediamine (EDA) and 2-propanol. Notably, samples containing higher proportions of Zr-$Ti_{0.3}Zr_{0.7}O_2$ and ZrO_2 produced EU in significant amounts, owing to their acid-base bifunctionality. Overall, EU was synthesized using CO_2-adsorbed $Ti_xZr_{(1-x)}O_2$ as a CO_2 source and as an accelerator for reacting with EDA, without requiring high-pressure or high-purity CO_2.

Keywords: CO2 capture and utilization (CCU) Ti-Zr mixed oxides TixZr(1−x)O2 Ethylene urea (EU) synthesis Acid-base bifunctionality

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