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## Triamine-Grafted Mesoporous Silica Materials for CO<sub>2</sub> Capture from the Atmosphere

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The concentration of CO<sub>2</sub>, the primary anthropogenic greenhouse gas (GHG) responsible for global warming and climate change, has experienced a rapid increase since the Industrial Revolution. The present levels of CO<sub>2</sub> surpass any recorded in human history, indicating a global average concentration of 425 parts per million by volume (ppmv) in 2024, as opposed to 315 ppmv in 1958. Additionally, there is a potential for it to reach 800 ppmv if multifaceted solutions are not implemented. This increase in emissions has caused a nearly 2-degree Fahrenheit rise in global temperatures since the pre-industrial era. CO<sub>2</sub> capture from the atmosphere, also known as direct air capture (DAC), might help us address this problem. The U.S. Department of Energy has endorsed DAC as one of the helpful strategies to achieve net-zero emissions by 2050. Owing to their superior performance, DAC applications involving cyclic adsorption-desorption of CO<sub>2</sub> by amine-modified silica materials (i.e., “aminosilica”) have gained momentum in recent years. These technologies have achieved significant attention because of their potential for commercialization and positive environmental impact to mitigate climate change. To that end, this project aims to develop materials suitable for DAC applications. This research involves the synthesis of triamine-grafted silica adsorbent materials. Triaminosilane was chosen for this purpose because it possesses two secondary amines and one primary amine, providing a high affinity for CO<sub>2</sub> capture. CARiACT G-10 silica (Fuji Silysia Chemical Ltd.) was used as support for the synthesis of aminosilicas. G-10 silica is commercially available at a low cost and has a high pore volume (1.2 cm<sup>3</sup>/g), large surface area (300 m<sup>2</sup>/g), small particle size (5 μm), and wide pores (20 nm), enabling it to attain high amine loadings and CO<sub>2</sub> uptakes, without compromising adsorption kinetics. Different materials were produced by implementing the grafting technique and varying factors such as the amount of water and triamine to synthesize unique samples. Using thermogravimetric analysis (TGA), the materials were screened for equilibrium CO<sub>2</sub> uptake, amine efficiency, and adsorption kinetics in the presence of dry CO<sub>2</sub> (400 ppmv, balance nitrogen) at 25 °C. One best-performing material with the highest CO<sub>2</sub> uptake and fastest CO<sub>2</sub> adsorption kinetics was chosen for rigorous 50-cycle testing under the above adsorption conditions, followed by regeneration at 120 °C in the presence of N<sub>2</sub>. The results indicated stable performance as evidenced by maintaining 99% of the initial CO<sub>2</sub> uptake throughout cycling. Using column-breakthrough testing, the performance of the final candidate was also evaluated in the presence of humid CO<sub>2</sub>, confirming previous reports that humidity boosts the CO<sub>2</sub> uptake of amine-modified materials. These results suggest the high potential of aminosilicas for DAC applications.

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