Chemical Engineering Doctoral Defense

Functional Materials for the Direct Air Capture of Carbon Dioxide

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Abstract

The excessive use of fossil fuels over the last few centuries have led to unprecedented changes in climate and a steady increase in the average surface global temperatures. Direct Air Capture(DAC) aims to capture CO2 directly from the atmosphere and alleviate some of the adverse effects of climate change. This dissertation focuses on methodologies to make advanced functional materials that show good potential to be used as DAC sorbents. Details on sorbent material synthesis and post-synthesis methods to obtain high surface area morphologies are described in detail. First, by incorporating K2CO3 into activated carbon (AC) fiber felts, the sorption kinetics were significantly improved by increasing the surface area of K2CO3 in contact with air. The AC-K2CO3 fiber composite felts are flexible, cheap, easy to manufacture, chemically stable, and show excellent DAC capacity and (de)sorption rates, with stable performance up to ten cycles. The best composite felts collected an average of 478 µmol of CO2 per gram of composite during 4 h of exposure to ambient (24% RH) air that had a CO2 concentration of 400-450 ppm over 10 cycles. Secondly, incorporating the amino acid L-arginine (L-Arg) into a poly(vinyl alcohol) (PVA) nanofiber support structure, created porous substrates with very high surface areas of L-Arg available for CO2 sorption. The bio-inspired PVA-Arg nanofiber composites are flexible and show excellent DAC performance compared to bulk L-Arg. The nanofiber composites are fabricated from an electrospinning process using an aqueous polymer solution. High ambient humidity levels improve sorption performance significantly. The best performing nanofiber composite collected 542 µmol of CO2 per gram of composite during 2 h of exposure to ambient, high humidity (100% RH) air that had a CO2 concentration of 400-450 ppm. Finally, poly(vinyl guanidine) (PVG) polymer was synthesized and tested for sorption performance. The fabrication of PVG nanofibers, divinyl benzene crosslinked PVG beads and glutaraldehyde crosslinked PVG were demonstrated. The sorption performance of the fabricated sorbents were tested with the glutaraldehyde crosslinked PVG having a dynamic sorption capacity of over 1 mmol of CO2 per gram of polymer in 3 h. The sorption capability of liquid PVG was also explored.

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