Chemical Engineering Doctoral Defense

Synthesis and Carbon Dioxide adsorption of Amine Modified Particulate Silica Aerogel Sorbents

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abstract

Post-combustion carbon capture is a viable option for reducing CO2 greenhouse gas emissions, and one potentially promising technology for this route is adsorption using chemically and physically based sorbents. A number of exceptional CO2 sorbents materials have been prepared including metal organic frameworks, zeolites, and carbon based materials. One particular group of capable materials are amine based solid sorbents that has shown to possess high adsorption capacities and favorable adsorption kinetics. A key variable in the synthesis of an amine based sorbent is the support which acts as the platform for the amine modification. Aerogels, due to their high porosities and surface areas, appear to be a promising support for an amine modified CO2 sorbent. Therefore, in order to develop a commercially viable CO2 sorbent, particulate aerogels manufactured by Cabot Corporation through an economical and proprietary ambient drying process were modified with amines using a variety of functionalization methods. Two methods of physical impregnation of the amino polymer TEPA were performed in order to observe the performance as well as understand the effects of how the TEPA distribution is affected by the method of introduction. Both samples showed excellent adsorption capacities but poor cyclic stability for lack of any covalent attachment. Furthermore the method of TEPA impregnation seems to be independent on how the polymer will be distributed in the pore space of aerogel. The last two methods utilized involved covalently attaching amino silanes to the surface silanols of the aerogel. One method was performed in the liquid phase under anhydrous and hydrous conditions. The materials developed through the hydrous method have much greater adsorption capacities relative to the anhydrous sample as a result of the greater amine content present in the hydrous sample. Water is another source of silylation where additional silanes can attach and polymerize. These samples also possessed stable cyclic stability after 100 adsorption/regeneration cycles. The other method of grafting was performed in the gas phase through ALD. These samples possessed exceptionally high amine efficiencies and levels of N content without damaging the microstructure of the aerogel in contrast to the liquid phase grafted sorbents.

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