RECOVERY OF MESOPOROUS SILICA AND VALUABLE CHEMICALS FROM THE RECYCLING OF SPENT SOLID CO_2 ADSORBENTS VIA CATALYTIC PYROLYSIS

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ABSTRACT

The Solid Adsorbent Looping Technology (SALT) is a promising CO_2 capture technology that utilizes strongly basic solid adsorbents, such as polyethyleneimine (PEI) supported on mesoporous silica (Si-PEI). Si-PEI circulates between a reactor for the adsorption of CO_2 from industrial flue gases and a regenerator to release the adsorbed CO_2 and regenerate the adsorbent. During operation, PEI gradually oxidizes, loses CO_2 adsorption capacity, and eventually must be replaced with fresh adsorbent. The commercial success of the technology depends on the reduction of the adsorbent replacement cost to <10€/ton CO_2 captured. In this work, we developed a technology for the recycling of the spent Si-PEI adsorbent via pyrolysis to a) recover the mesoporous silica for reuse in the production of fresh Si-PEI and b) recover valuable chemicals from the thermal decomposition of PEI to generate additional revenue.

Spent Si-PEI adsorbent from a previous work [1] was pyrolyzed at 400-650 °C to thermally decompose the oxidized PEI and recover the silica. At ≥500 °C, near-complete degradation of PEI was achieved, and silica with a pore volume of ca. 1.2 cm^3/g was obtained, i.e., about 70% of the virgin silica's pore volume. The characterization of the pyrolysis oils revealed the presence of alkyl-pyrazines, heteroaromatics compounds with many applications in the food, fragrance, and pharmaceutical industries [2]. A method based on gas chromatography was developed to quantify the alkyl-pyrazines, whose yield was determined to range from 1.7 wt.% at the lowest pyrolysis temperature (400) °C to 4.9 wt.% at the highest (650 °C). The upgrading of the PEI pyrolysis vapors with different catalysts was studied to maximize the alkyl-pyrazines; using an ecat FCC catalyst at a Si-PEI:catalyst ratio of 1:1, an alkyl-pyrazine yield of 7 wt.% was achieved at a pyrolysis temperature of 600 °C. The process was upscaled to produce recovered silica for reimpregnation with fresh PEI. Larger-scale runs confirmed the pore volume of 1.2 cm^3/g of the recovered silica, on which a 43 wt.% loading of fresh PEI was achieved, just slightly lower compared to the 47 wt.% PEI loading that was achieved on virgin silica.

KEYWORDS: Recycling, Mesoporous silica, Pyrazines, CO_2 adsorbents, CO_2 capture

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