Ga-Substituted Hydrotalcites Capture CO_2 at Elevated Temperatures

Fossil fuel combustion for power generation currently accounts for 40% of all CO_2 emissions. Newly constructed coal-fired power plants are expected to number about 100 per year for the next 25 years, resulting in doubling current CO_2 emissions. With the global carbon cycle now unbalanced by 3.2 gigatons of excess CO_2, the capture of CO_2 from power-plant emissions is imperative. Known CO_2 sorbents are either easily depleted or would bring an 8–35% energy penalty due to regeneration requirements. New CO_2 sorbents must overcome these limitations and must also operate at the elevated temperatures of power-plant operation. Hydrotalcites (HTs) are layered double hydroxide salts with the following stoichiometry:

$M_{1-x}^{2+}M_x^{3+}(OH)_2A_{x/m}^{m-}\cdot yH_2O$

where the divalent ion is typically Mg^{2+}, the trivalent ion Al^{3+}, and the anion is Cl^-, NO_3^-, or CO_3^{2-}. HTs are known to be stable adsorbents even under harsh conditions that have been previously reported to capture CO_2. Recently, however, substituted HTs (SHTs) with stable and reversible CO_2 adsorption and markedly higher capture capacity than unsubstituted HTs have been reported by the research team C.T. Yavuz, P.C. Ford, and G.D. Stucky at the University of California, Santa Barbara; A.V. Iretskii of UCSB, Georgia Institute of Technology, and Lake Superior State University; B.D. Shinall at Georgia Tech; M.G. White at Georgia Tech and Mississippi State University; T. Golden at Air Products and Chemicals; and M. Atilhan at UCSB and Qatar University.

As reported in the August 11 issue of *Chemistry of Materials* (DOI: 10.1021/cm900834g ; p. 3473), the researchers modified a conventional procedure for synthesizing HT to make Ga-substituted HTs (Ga-SHT; 10 mol-% Ga substituted for Al). In another modification, a portion of the material was stirred into 2M K_2CO_3 before removing the water. The SHTs were then converted to CO_2 sorbents by calcining at 400°C in an inert atmosphere. The capacity to adsorb CO_2 was measured using gravimetric analysis at 200°C and a CO_2 partial pressure of 0.7 atm CO_2 in a dry, inert gas stream. K_2CO_3-promoted, calcined HT and calcined Ga-SHT both showed an adsorption capacity of 1.3 mmol of CO_2 adsorbed per gram of adsorbent, which is comparable to the industrial standard for CO_2 removal, monoethanol amine. Exchanging the HTs with potassium carbonate leads to calcined materials with about a three-fold increase in CO_2-adsorption capacity. Several adsorption-desorption cycles were performed by changing the temperature from 200°C to 400°C and back. The Ga-substituted HTs proved to be the most robust, maintaining their CO_2 adsorptivity for at least three cycles, with 10% Ga substitution found to be best. Powder x-ray diffraction studies showed that the HT framework of Ga-SHTs is substantially preserved even after three cycles, and that SHTs were more quickly regenerated from calcined solids when exposed to moist air.

The researchers said that their work "demonstrates that substituted hydrotalcites are viable options as efficient and selective CO_2 sorbents in the presence of other components in the gas stream including water, hydrocarbons, and carbon monoxide."

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