

**Manipulation of CaCO<sub>3</sub>  
crystallization in mineral  
carbonation of flue gas  
desulfurization gypsum**

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Aqueous ammonia solution containing flue gas desulfurization (FGD) gypsum, a byproduct obtained from coal-fired power plants, was used in direct aqueous mineral carbonation for CO<sub>2</sub> sequestration. Previously, we found the presence of induction period during which highly supersaturated CaCO<sub>3</sub> exists as solvated forms in advance of CaCO<sub>3</sub> crystallization process. In this work, we attempted to manipulate the crystallization of high-purity CaCO<sub>3</sub> varying experimental parameters such as ammonia content or introducing polyacrylic acid (PAA) as an inhibitor for CaCO<sub>3</sub> crystallization. We particularly focused on polymorphs, morphologies, and yields of not only dissolved CaCO<sub>3</sub> but also consequently formed precipitated CaCO<sub>3</sub>. The amount of dissolved CaCO<sub>3</sub> was found to increase in proportion to the ammonia content because of carbamate ions resulting from excess ammonia. With the addition of 2.7 g/L PAA, the amount of dissolved CaCO<sub>3</sub> increased from 5 wt.% to more than 60 wt.%. The evolution sequence of polymorphs of CaCO<sub>3</sub> was examined using an in-situ liquid-phase X-ray diffraction using the solution filtered during the induction period. In the absence of PAA, the initially formed amorphous CaCO<sub>3</sub> transformed to round-shaped vaterite, and then to the typical rhombohedral calcite after exposure to water. In the presence of PAA, the initially formed amorphous phase turned directly into calcite phase. The crystal morphologies varied from twinned pseudo-spherical to dumbbell-shaped depending on the amount of PAA injected. These findings demonstrate that the precipitation of pure CaCO<sub>3</sub> can be manipulated to a large extent exploiting the induction period.