

Mitigating Environmental Impacts of CO₂ Through Mineralization Using FGD Gypsum

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The IPCC (2019) has stated that CO₂ emission reductions alone will not be sufficient to prevent a 2°C increase in mean global temperatures by 2050. Steps must also be taken to reduce atmospheric CO₂ concentrations through carbon capture and storage. Methods of carbon storage include geologic/ocean sequestration and CO₂ mineralization. The focus of this study is CO₂ mineralization.

Prior studies aimed at CO₂ mineralization have proven effective for long-term, stable storage of CO₂ but were prohibitive due to slow reaction rates and/or high energy costs. More recent mineralization studies reacting flue gas desulphurization (FGD) gypsum (CaSO₄·2H₂O) with CO₂ in alkaline solutions, have shown the conversion of gypsum into calcium carbonate (CaCO₃) to be a rapid and effective method for carbon storage. However, most of these studies used ammonia or ammonium hydroxide to increase solution pH levels. This method is inefficient because the Haber process used to generate ammonia requires elevated temperatures (400-500°C), and is a significant source of CO₂.

The goal of this study was to obtain high FGD gypsum-to-calcite conversation rates at ambient temperature while eliminating the need for ammonia. A stirred reactor was utilized to study the effects of P_{CO2} (0.6895, 2.0684, 4.1369, 17.2369 bar), solution pH (12, 13, 13.5 and 14), solid:liquid ratio (1:10, 1:40, 1:80, 1:100), and reaction time (10, 15, 30, and 120+ min) variation on the extent of carbonation.

Experimental results showed solution pH was a primary control on mineralization, with nearly 100% conversion of FGD gypsum to CaCO₃ occurring at initial pH=13.5 and 14, for P_{CO2}>2 bar and S:L=1:100. No gypsum conversion to CaCO₃ resulted at initial pH=12, regardless of the P_{CO2} and S:L used. Time also played a significant role in the extent of gypsum carbonation in the time-series experiments performed at initial pH=13. The optimum reaction time was 15 min for P_{CO2}=2.068 bar and S:L=1:100, achieving 75% conversion. In contrast, a reaction time of 360 min resulted in only 61% net conversion at the same P_{CO2} and S:L ratio, due to pH dropping below 7. The S:L ratio used also controlled the extent of gypsum conversion to CaCO₃, with decreasing S:L ratio resulting in higher percent conversion values. The results of this study demonstrate that FGD gypsum can serve as a viable feedstock for CO₂ mineralization, potentially providing a rapid and inexpensive method for carbon storage.