

Supplementary Information

Evolution of Silicate Coordination in Architected Amorphous and Crystalline Magnesium Silicates during Carbon Mineralization

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Table S1. Mean particle sizes of the unreacted and carbonate-bearing silicate sorbents.

Mg-silicates	Unreacted	H₂O, Stirring	NaHCO₃, Stirring	H₂O, No-stirring	NaHCO₃, No-stirring
AC	13.05 μm	99.15 μm	31.95 μm	14.03 μm	9.61 μm
HC	25.04 μm	15.78 μm	8.01 μm	14.32 μm	7.99 μm

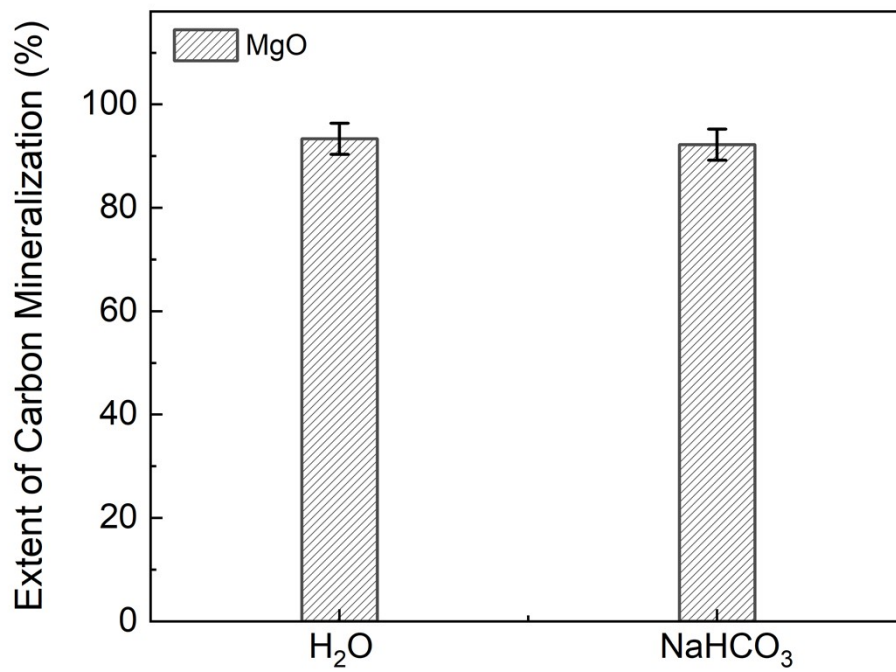


Figure S1. The carbon mineralization extent of MgO without stirring at 200 °C, 20 atm.

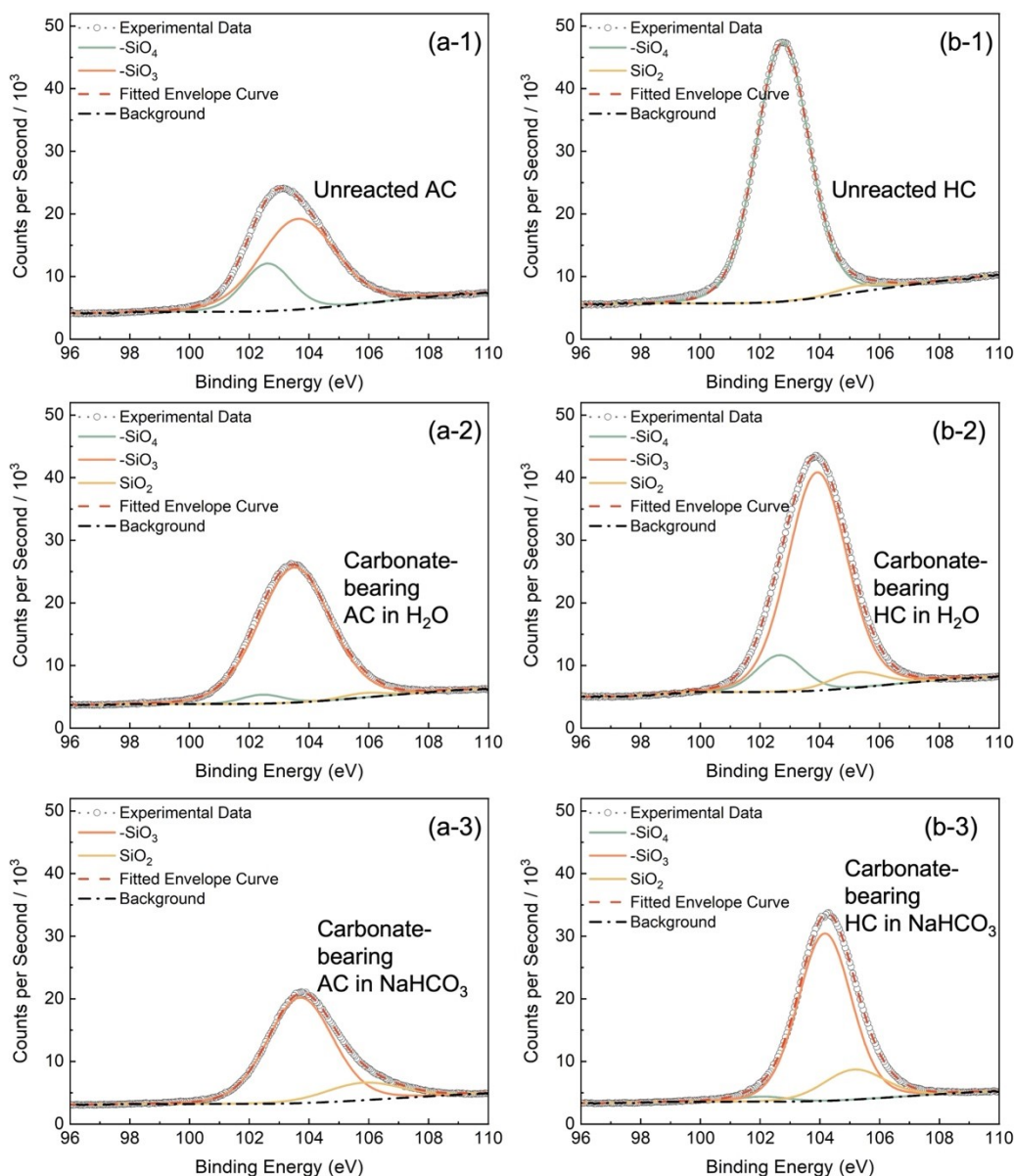


Figure S2. The deconvoluted Si2p spectrum of the (a-1) AC Mg-silicate, (a-2) carbonate-bearing AC Mg-silicate with DI-water, (a-3) carbonate-bearing AC Mg-silicate with NaHCO₃ and (b-1) HC Mg-silicate, (b-2) carbonate-bearing HC Mg-silicate with DI-water, (b-3) carbonate-bearing HC Mg-silicate with NaHCO₃, in stirring mode at 200 °C, 20 atm determined by X-ray photoelectron spectroscopy.

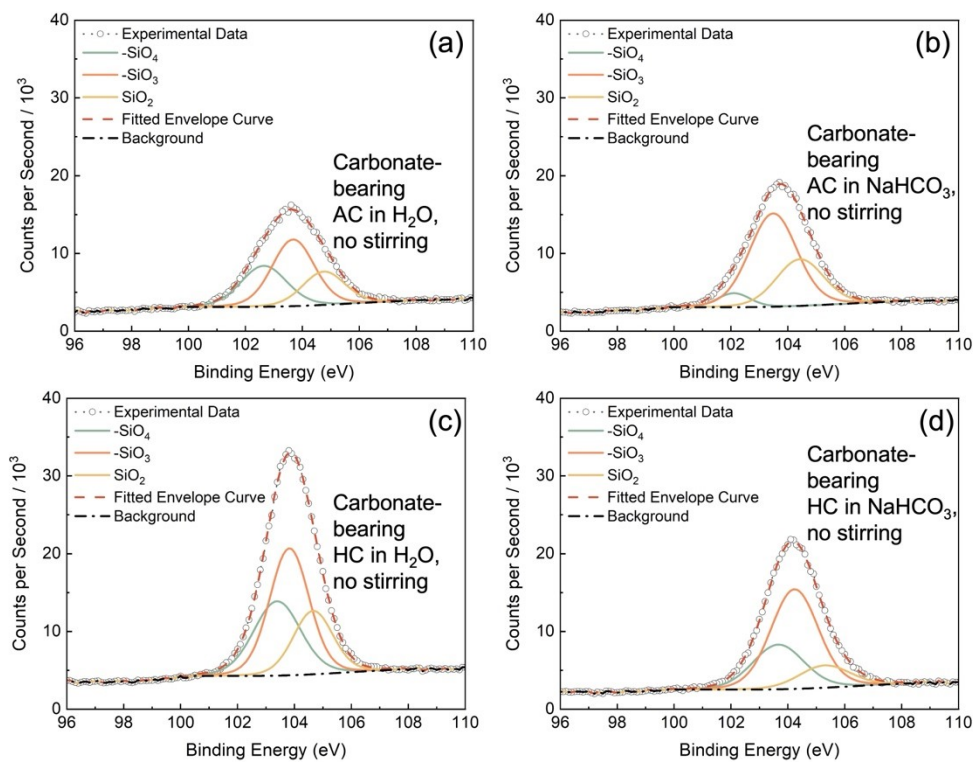


Figure S3. The deconvoluted Si_{2p} spectrum of the (a) carbonate-bearing AC Mg-silicate with DI-water, (b) carbonate-bearing AC Mg-silicate with NaHCO₃, (c) carbonate-bearing HC Mg-silicate with DI-water, and (d) carbonate-bearing HC Mg-silicate with NaHCO₃, in no-stirring mode at 200 °C, 20 atm determined by X-ray photoelectron spectroscopy.

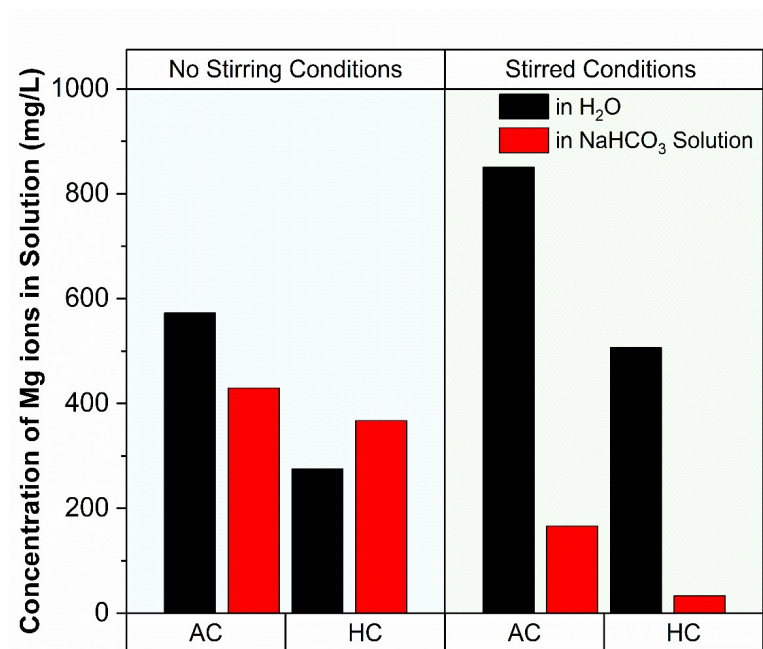


Figure S4. The concentration of Mg ions in the collected aqueous samples determined by Inductively Coupled Plasma (ICP) analysis.

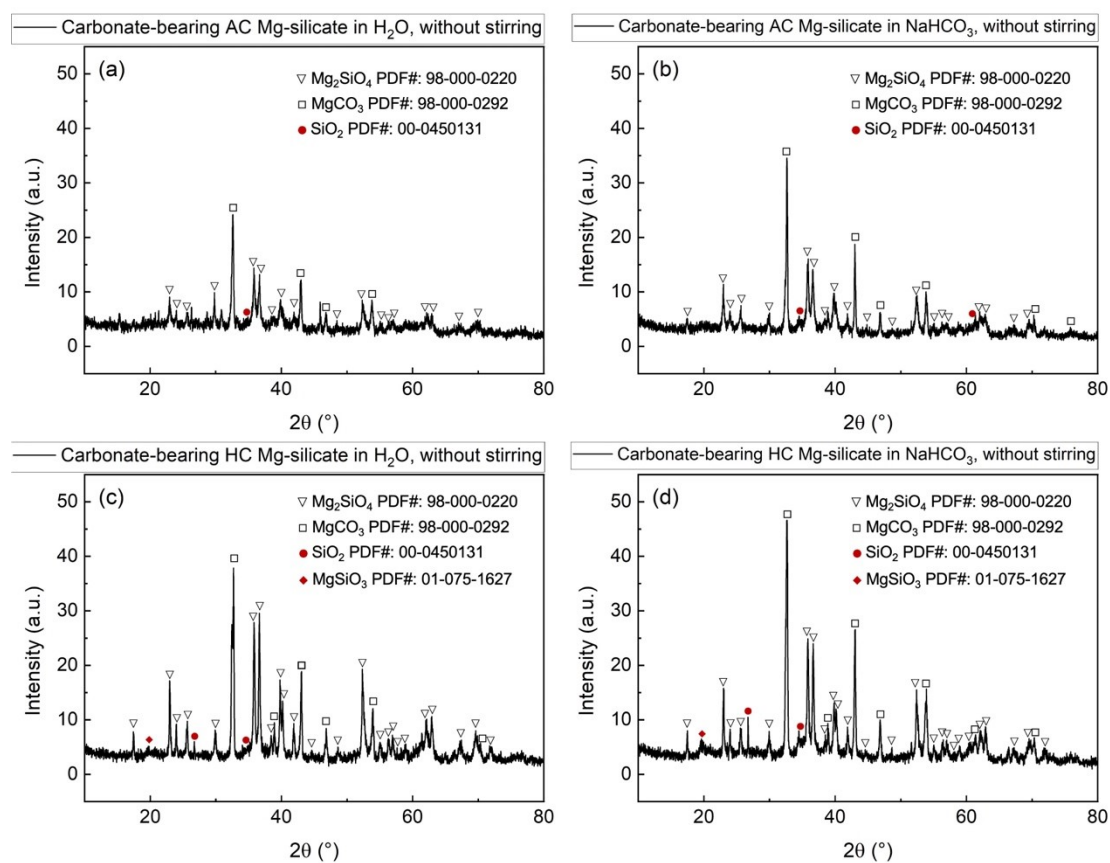


Figure S5. Structural arrangement of (a) carbonate-bearing AC Mg-silicate with DI-water, (b) carbonate-bearing AC Mg-silicate with $NaHCO_3$, (c) carbonate-bearing HC Mg-silicate with DI-water, and (d) carbonate-bearing HC Mg-silicate with $NaHCO_3$ in no-stirring mode at $200\text{ }^{\circ}C$, 20 atm determined by X-ray diffraction measurements.

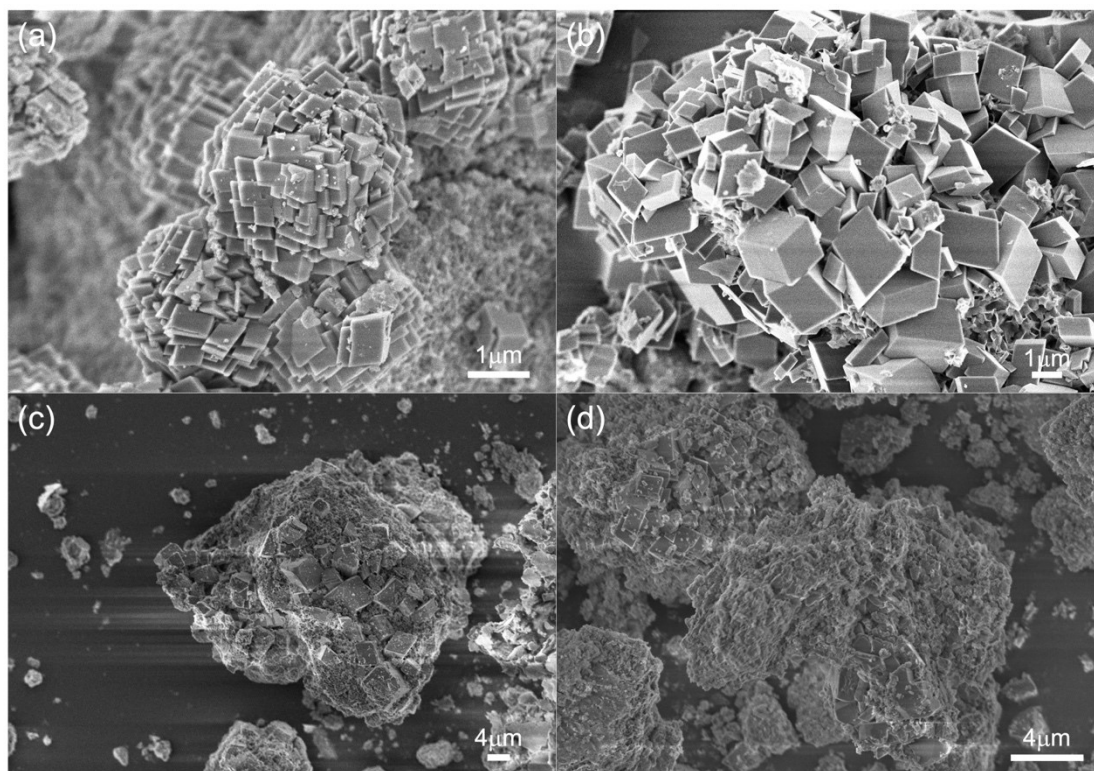


Figure S6. Morphologies determined using Scanning Electron Microscopy (SEM) of: (a) carbonate-bearing AC Mg-silicate with DI-water, (b) carbonate-bearing AC Mg-silicate with NaHCO_3 , (c) carbonate-bearing HC Mg-silicate with DI-water, and (d) carbonate-bearing HC Mg-silicate with NaHCO_3 in no-stirring mode. Experiments are conducted at 200 °C, $p\text{CO}_2$ of 20 atm for a reaction time of 3 hours.