Supporting Information

Unveiling the Critical Process in Which Organic Molecules Control the Polymorphism of Magnesium-Containing Calcium Carbonate: the Early Nucleation of Amorphous Precursors or the Subsequent Amorphous to Crystalline Transformations?

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**Fig. SI-1** Raman spectra of (A) ACC, (B) SC-ACC, (C) PEO-ACC and (D) PEI-ACC precursors. The broad peak at 1085 cm$^{-1}$ is the characteristic peak of amorphous calcium carbonate.$^{[1]}$
Fig. SI-2  XRD pattern of the samples crystallized from ACC precursor after 24 h of reaction in various redispersion media: (A) DI water; (B) trisodium citrate; (C) sodium alginate; (D) PEO; and (E) PEI aqueous solution.
**Fig. SI-3** XRD patterns of the samples crystallized from amorphous precursors formed in the presence of various concentrations of SC after 24 h of reaction in DI-water.
Fig. SI-4  XRD patterns of the samples crystallized from PEI-ACC precursors after 48 h of reaction in DI water, trisodium citrate, sodium alginate, and PEO aqueous solution, respectively.
Fig. SI-5  XRD patterns of the samples crystallized from amorphous precursors prepared in the presence of ethylene glycol (1 mL) and hydrazine (1 mL), in place of the high MW polymers, PEO and PEI, respectively, after 48 h of reaction in DI water.
Fig. SI-6  XRD patterns of the samples crystallized from ACC, SC-ACC, PEO-ACC and PEI-ACC precursors dispersed in DI water for 7 days without the dispersion process via sonication.