## **Supporting Information**

## Naturally Occurring and Biomimetic Synthesized Calcite Spherulites

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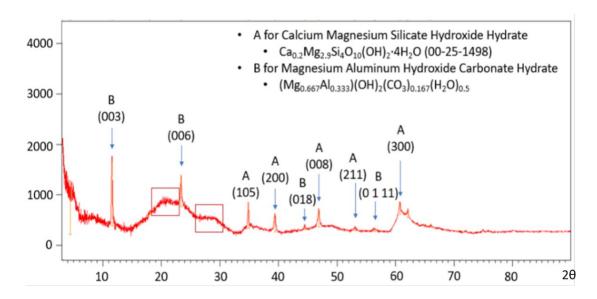
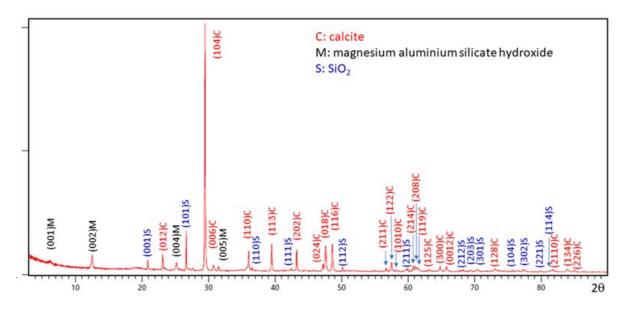
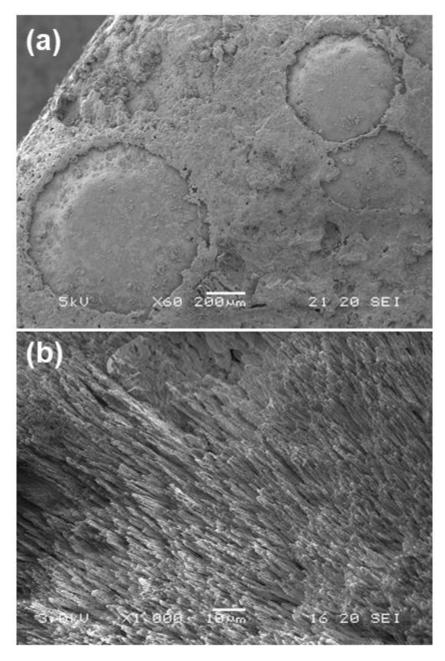


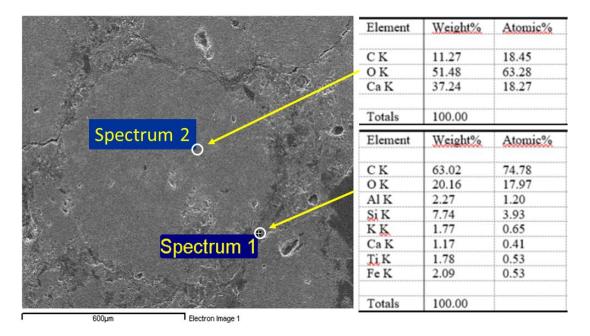
Figure S1. XRD pattern from a dried stevensite specimen.



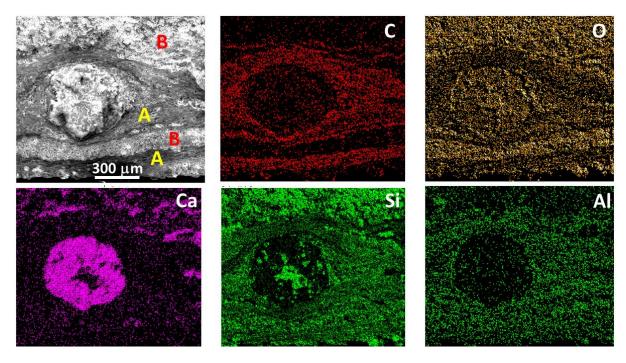
**Figure S2.** A typical XRD pattern from natural rock sample containing many calcite spherulites.



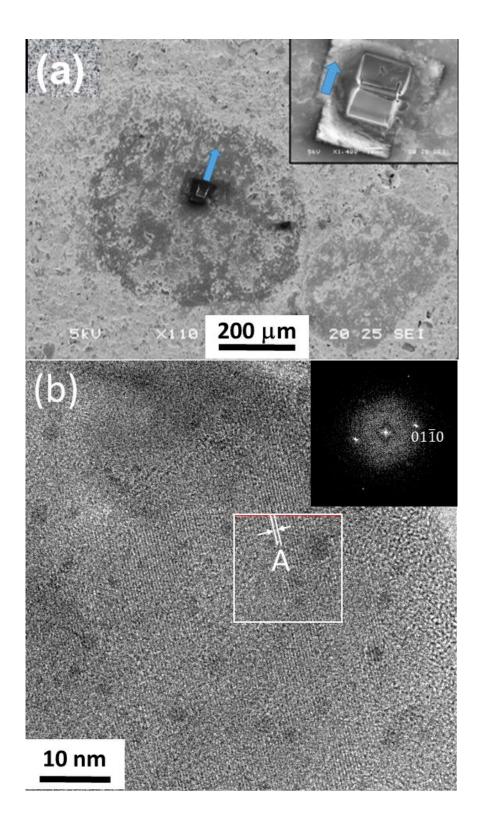
**Figure S3.** (a) SEM image of a rock sample with two spherulite exposing to the surface. (b) SEM image of a spherulite surface after acid treatment, revealing the microrods.



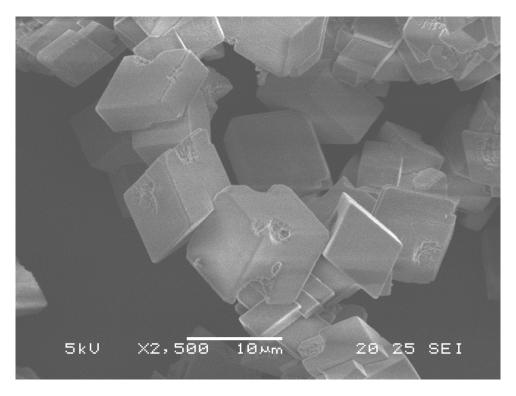
**Figure S4.** EDX point analysis of a spherulite with a thin C-rich surface layer.



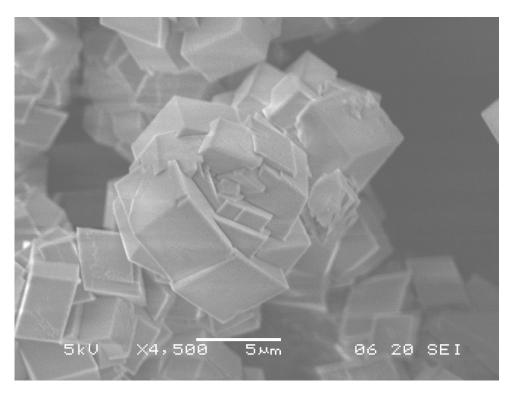
**Figure S5.** EDX elemental mapping of a spherulite with its surroundings. Top-left picture is a SEM image of the area. The spherulite is covered by two layers, A is C-rich and O-poor. B is mainly silicate.



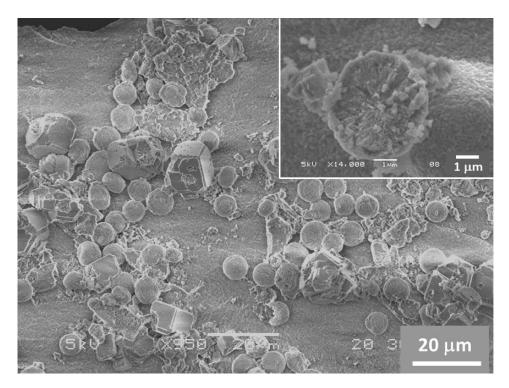
**Figure S6.** (a) SEM image of a naturally occurring spherulite showing the location of FIB cutting. The resulted specimen plate is normal to the radial direction. (b) The corresponding HRTEM image of an edge of a microrod, showing many separated nanocrystallites. The fringes "A" are indexed to (01-10) of the calcite structure.



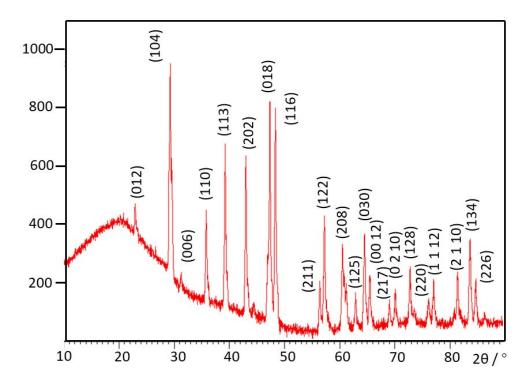
**Figure S7.** SEM image of rhombohedral crystals in group 1 via a simple mixture of 5.0 mmol  $CaCl_2$  and  $Na_2CO_3$  solutions. The reaction time is 7 days.



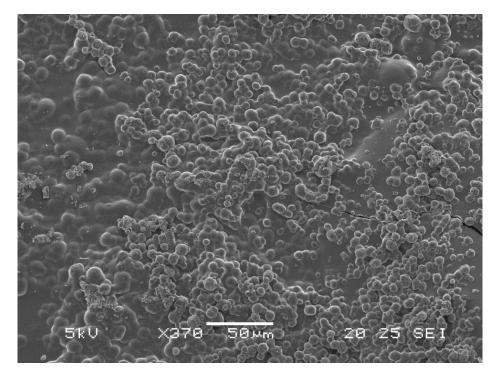
**Figure S8**. SEM image of rhombohedral crystals in group 2 via a mixture of  $5.0 \text{ mmol CaCl}_2$  and a stevensite suspension, followed by addition of  $5.0 \text{ mmol Na}_2\text{CO}_3$  solution. The reaction time is 72 h.



**Figure S9**. SEM image of a specimen in Group 4 with reaction time of 3 h, showing a mixture of bulk crystals and spherulites. The inset is a SEM image of cross section of a spherulite with the reaction time of 30 min.



**Figure S10**. PXRD pattern of Group 6 sample with reaction time of 30 min. The diffraction peaks are indexed to the rhombohedral structure of calcite. The sample also contains a large amount of alginate.



**Figure S11**. SEM image of a specimen in Group 7 with reaction time of 6 days, showing a high stability of the spherulites.

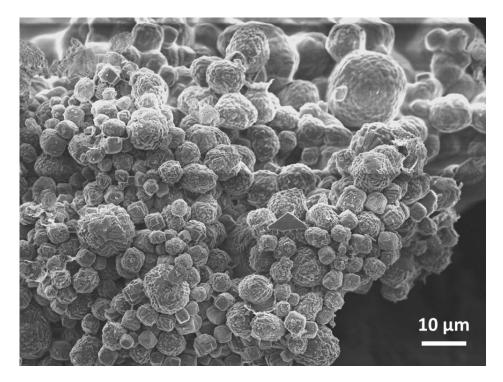


Figure \$12. SEM image of a specimen in Group 8 with a reaction time of 1 h.

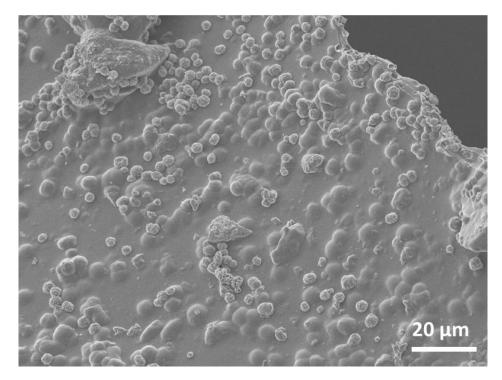
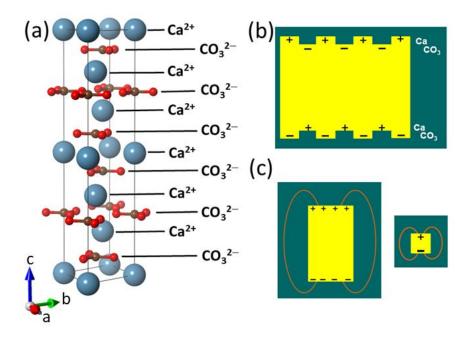


Figure \$13. SEM image of a specimen in Group 9 with a reaction time of 1 h.



**Figure S14**. Schematic drawing of (a) the structure of calcite, showing alternative layers of  $Ca^{2+}$  and  $CO_3^{2-}$  along the c axis, (b) a large crystal with uneven {0001} surfaces, (c) large and small crystals with charge separation to form a dipolar field.