Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Calcite crystallization in the cement system: morphological diversity, growth mechanism and shape evolution

Supportive Information

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1. Preparation for C_3S

Pure tricalcium silicate (Ca_3SiO_5, C_3S) was synthesized from a 3: 1 stoichiometric mixture of $CaCO_3$ and SiO_2. The powders were mixed uniformly for 24 hours and dried at 100 °C for 24 hours. Afterwards, the mixture was pressed into pellets and heated at 1600 °C for 12 hours with a 200 °C per hour rate. After 6 hours, the heating process was interrupted to quench the sample in air. Then, the pellets were reground and recompacted for further heating for three times.

2. Some properties for C_3S

The particle size distribution of C_3S was determined with the aid of laser particle size analyzer, shown in Figure S1.

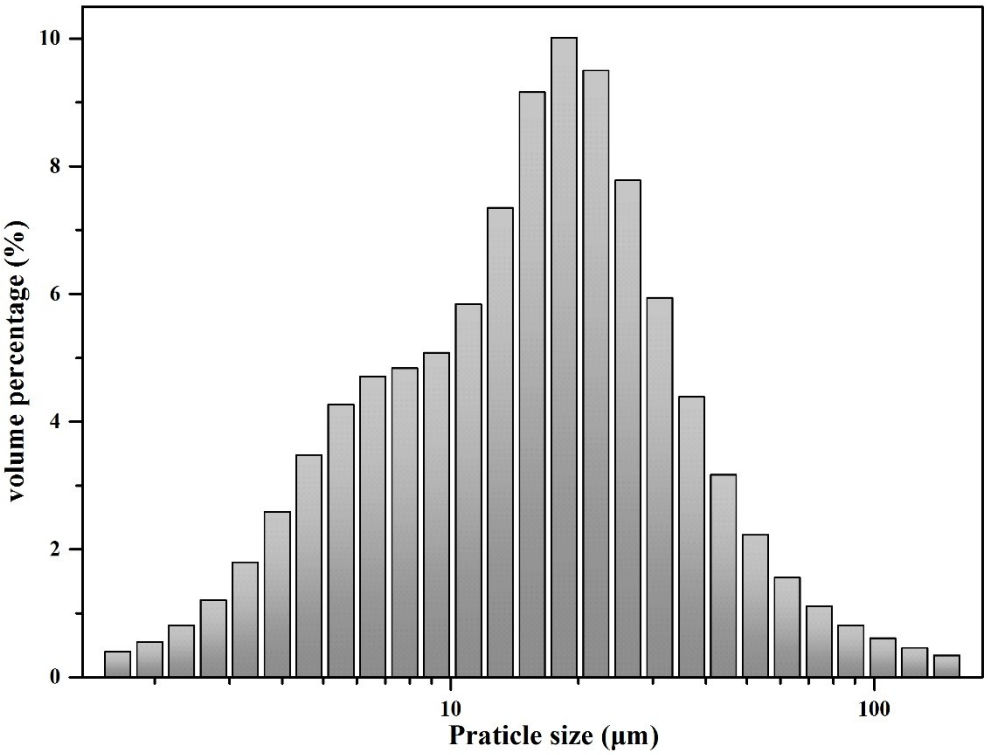

Figure S1 Particle size distribution of C_3S.

The purity and crystalline form were examined with XRD (X–ray diffraction) in Figure

S2.

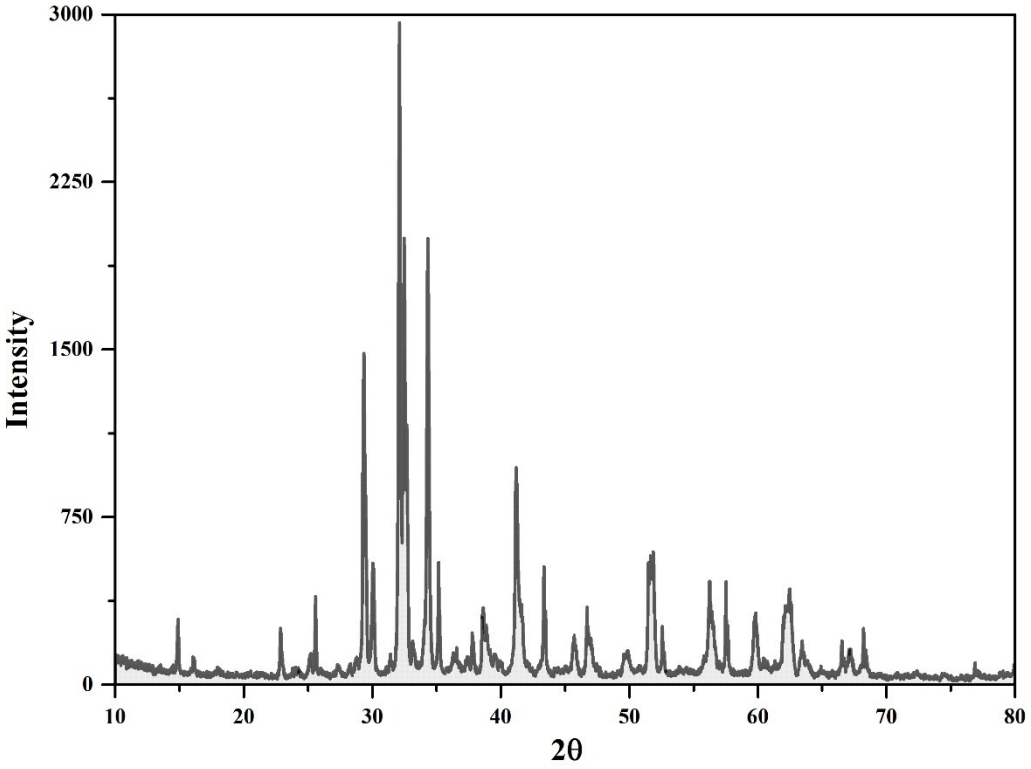

Figure S2 XRD pattern of synthesized C_3S.

3. Preparation for calcium silicate hydrate

Solutions of calcium nitrate, sodium silicate and sodium hydroxide were prepared in decarbonized water according to the Kumar [1]. To be detail, C–S–H was synthesized by gradually adding calcium nitrate solution (1 $mol \cdot L^{-1}$) with continual stirring under nitrogen atmosphere to sodium silicate solution. The nominal Ca/Si molar ratio was 1.7 for C–S–H. Notably, pH value was kept between 13.1 and 13.3 by adding NaOH during the precipitation of C–S–H.

4. SAED pattern for calcium silicate hydrate

4.1 Calcium silicate hydrate prepared by diluted hydration of C_3S

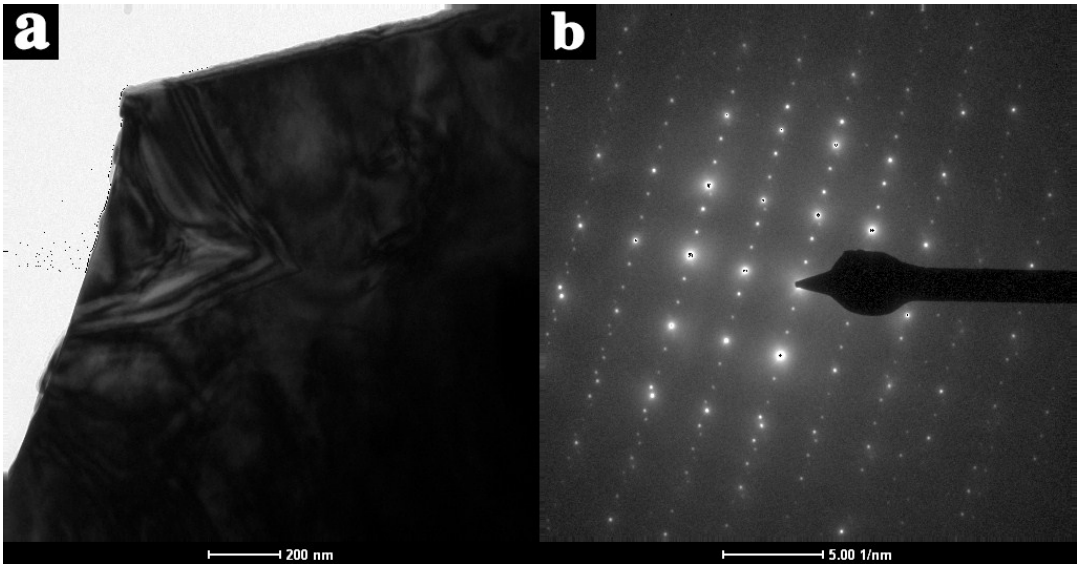

Figure S3 (a) Morphology of C_3S and (b) its SAED pattern.

Figure S3 exhibits the morphology of C_3S, which is indicative of prefect crystals with clear electron diffraction.

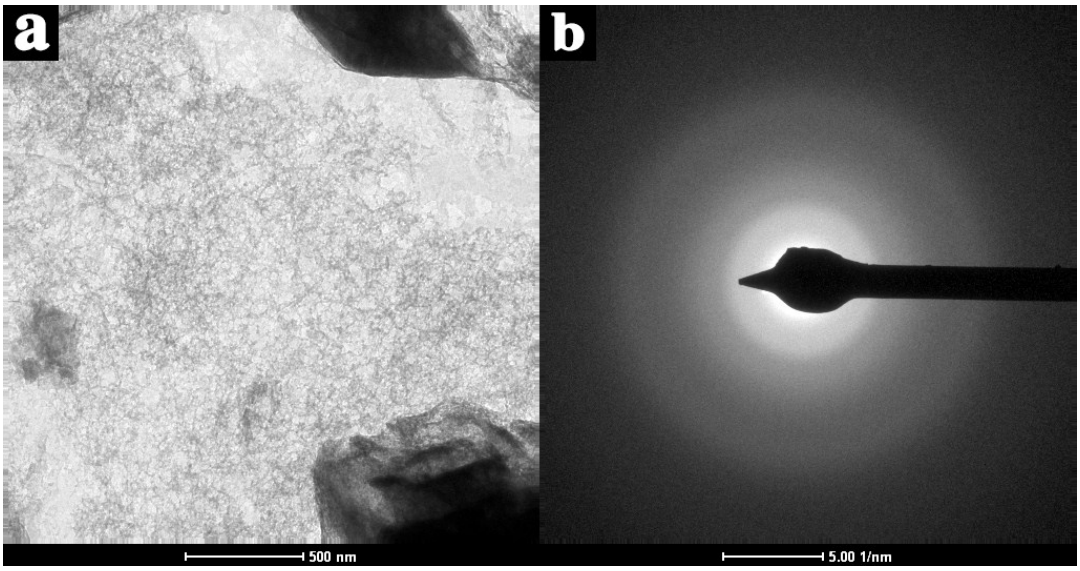

Figure S4 (a) Morphology of calcium silicate hydrate obtained by diluted hydration and (b) its SAED pattern.

After 12 hours' hydration, some amorphous phases are found with wide rings shown in Figure S4. This kind of phase is determined to calcium silicate hydrates, which is coincide with the results in literature.

4.2 Synthesized calcium silicate hydrate

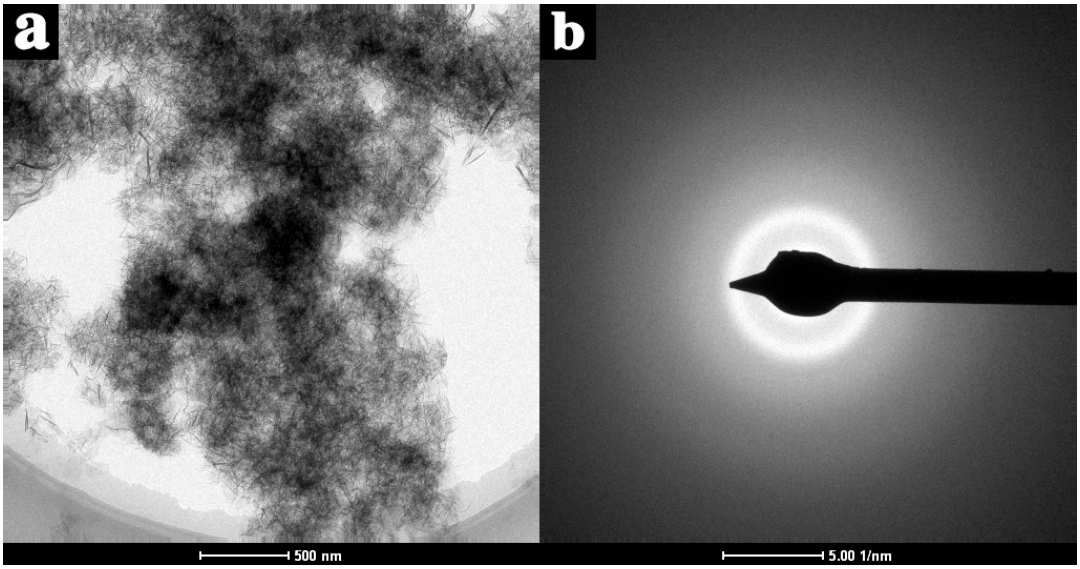

Figure S5 (a) Morphology of synthesized calcium silicate hydrate and (b) its SAED pattern.

The artificial calcium silicate prepared in the aforementioned procedures are determined to be amorphous as well, coincide with the results obtained by Kumar [1].

4.3 Particle size distribution measurement

The particle size distribution is processed by the software, Nano Measure. As shown in Figure S6, we measured the size according to the SEM images. Admittedly, there remains the problem of aggregation of particles and we analyze the figures by artificial adjustments (like 801 particle in Figure S6) to eliminate this problem. Although the particle size distribution is not totally correct, it could still provide some quantitative or semi quantitative results.

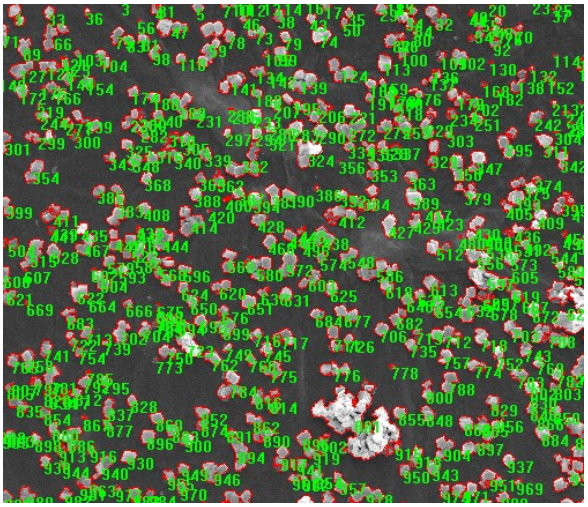

Figure S6 A method to determine the size distribution.

References

(1) Kumar, A.; Walder, B. J.; Kunhi Mohamed, A.; Hofstetter, A.; Srinivasan, B.; Rossini, A. J.; Scrivener, K.; Emsley, L.; Bowen, P. The Atomic-Level Structure of Cementitious Calcium Silicate Hydrate. *J. Phys. Chem. C* **2017**, *121* (32), 17188–17196.