Supplementary information:

1. Phase characterization (XRD)

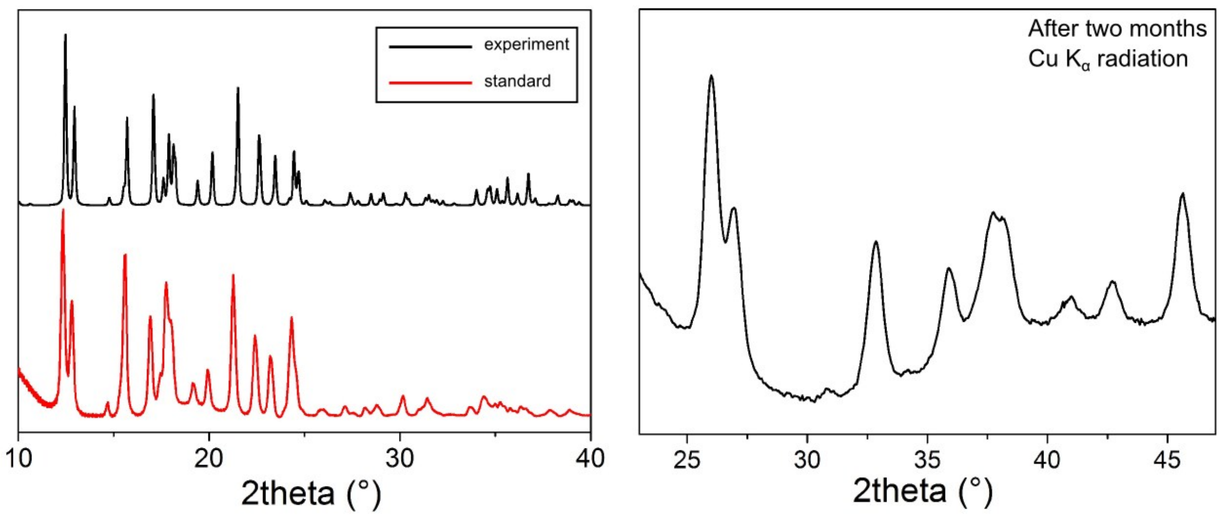

Fig. S1 Left: X-ray diffraction patterns (synchrotron) of silica aragonite biomorphs (black) and standard aragonite (red); Right: X-ray diffraction pattern (Cu K_{α}) of silica aragonite biomorphs stored at room temperature after two months.

2. Phase characterization (Raman spectroscopy)

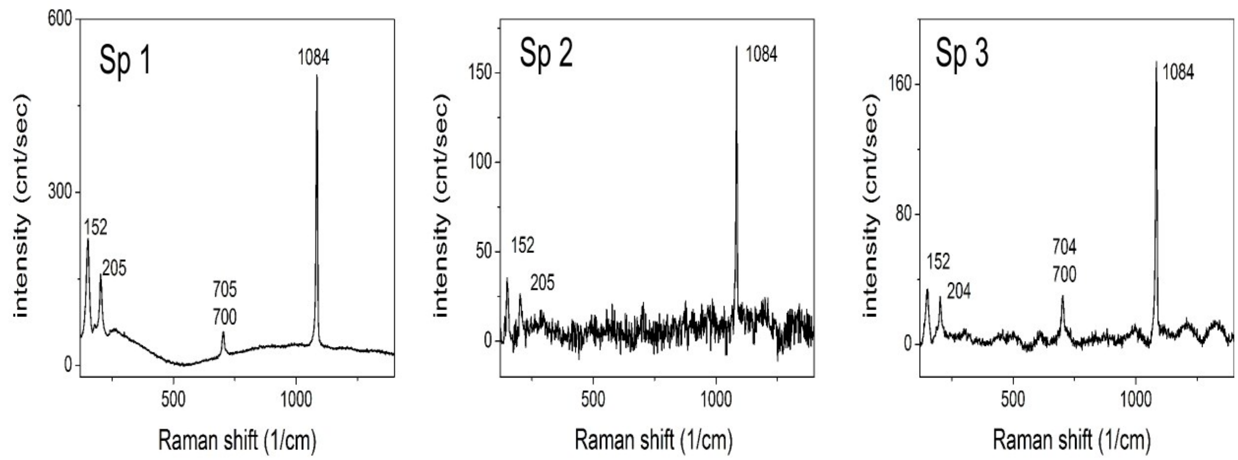

Fig. S2 Raman spectra of silica aragonite biomorphs: (Sp 1) the spectrum of diabolo-like core (acquisition time: 15 s per run); (Sp 2) the spectrum of the rim of the petal-like laminar sheet (acquisition time: 30 s per run); (Sp 3) the spectrum of the center of the petal-like laminar sheet (acquisition time: 15 s per run).

3. Analysis of chemical composition (EDX)

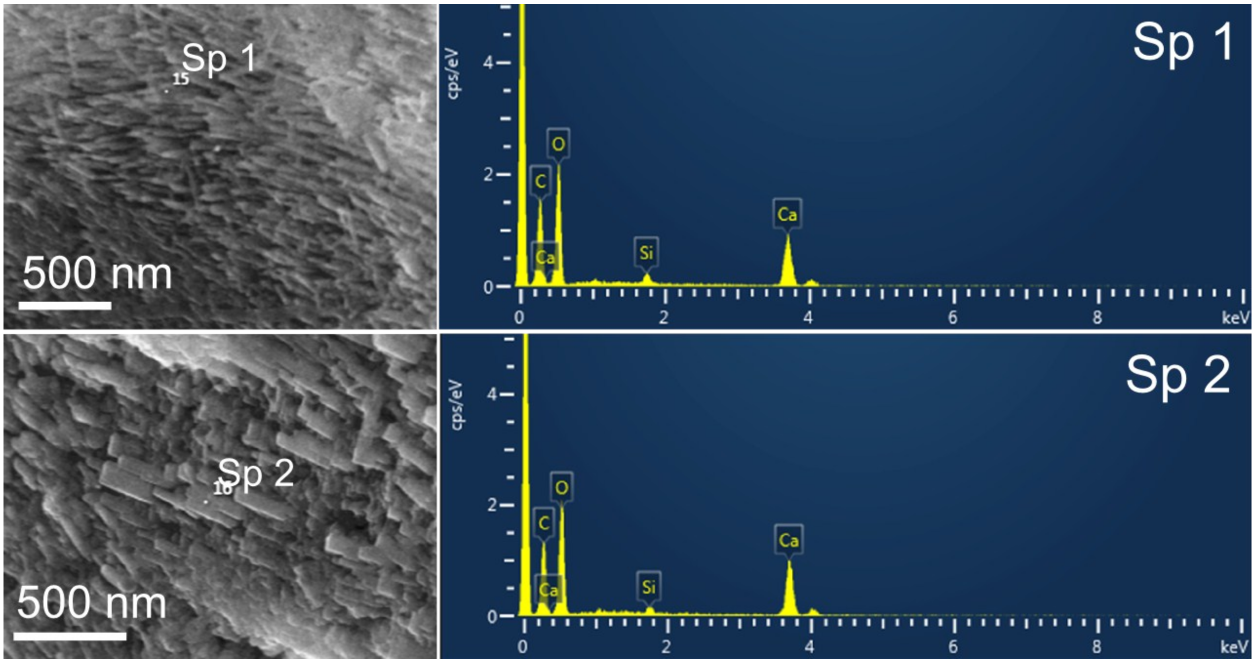

Fig. S3 EDX spectra collected from different positions of flower-like particle of silica aragonite biomorphs: (Sp 1) spectrum collected from the surface of the petal-like laminar sheet; (Sp 2) spectrum collected from the dumbbell-shaped core of the flower-like particle. The precise locations of EDX were labelled by white dots in the FESEM images.

4. Experiment at higher temperatures (45, 60 and 80 °C)

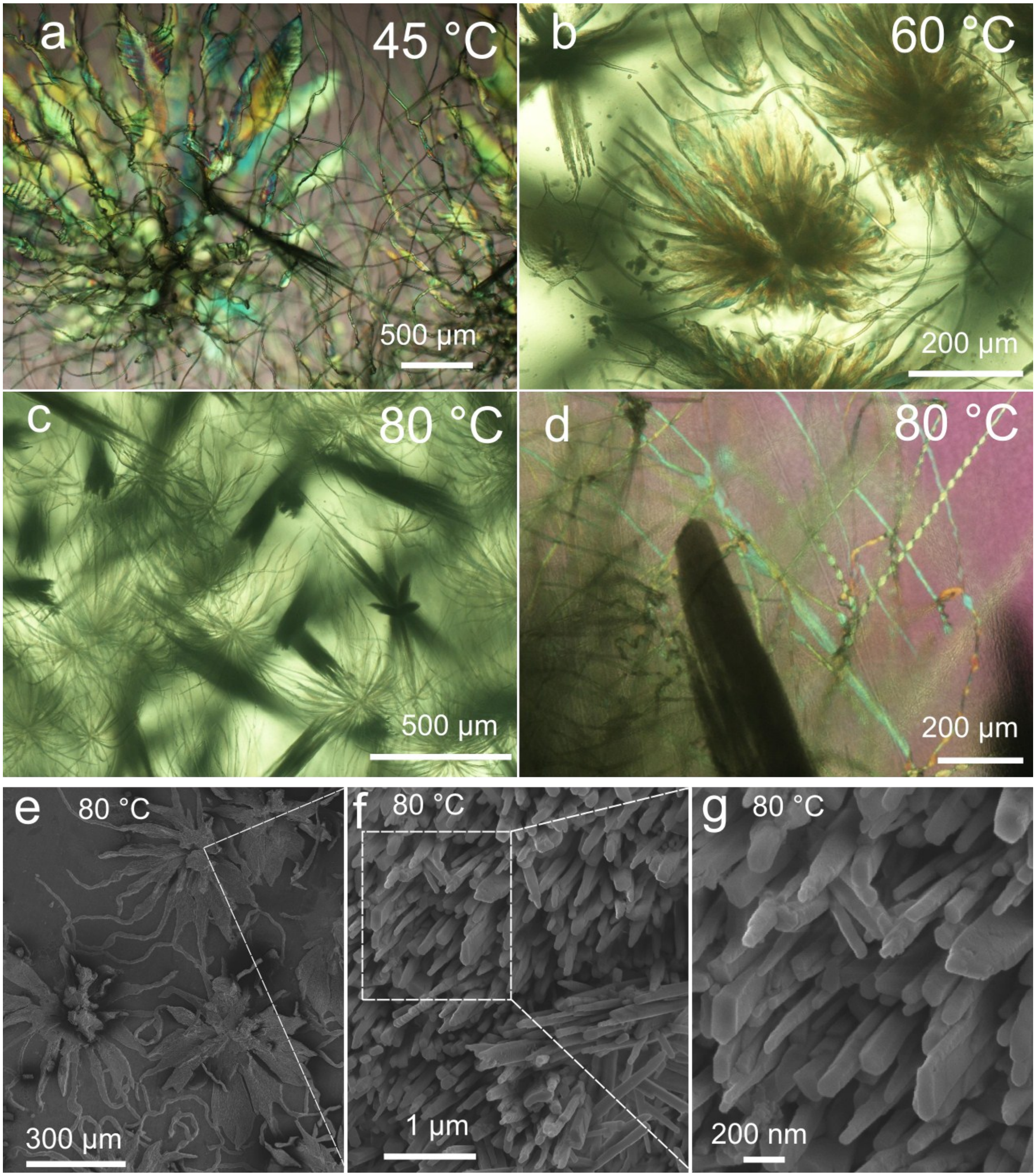

Fig. S4 Optical micrographs of the various silica aragonite biomorphs formed at higher temperatures: (a) 45 °C; (b) 60 °C; (c and d) 80 °C; and the FESEM micrographs of aragonite biomorphs formed at 80 °C (e, f, g). The dumbbell-shaped core of the particle was smaller at higher temperatures, and the petal-like laminar curving sheets became larger and longer (a, b, c and e). Due to the continuous curling on the rims, the laminar sheet was twisted and further developed into tentacle-like filaments (a, b, d and e) or helical filaments (d). More aragonite formed at higher temperatures, but the appearance of fibrous calcite could not be completely inhibited (dark shadows in a, c and d). The silica gel was usually difficult to be cleaned totally from the surface of the biomorphic particles due to the complex shape and the fragility, and this residual silica gel hinder the texture study on the surface, especially for the particles with more tentacle-like filaments formed at higher temperatures. From the cross-section of the fragment where the actual particles were exposed, the FESEM micrograph revealed that the biomorphic particles also consisted of densely packed co-oriented nanorods (f); and close-up view (g) indicates these nanorods were pseudo-hexagonal crystals which were the same to the nanorods formed at room temperature.

5. FESEM images of aragonite biomorphs formed at room temperature

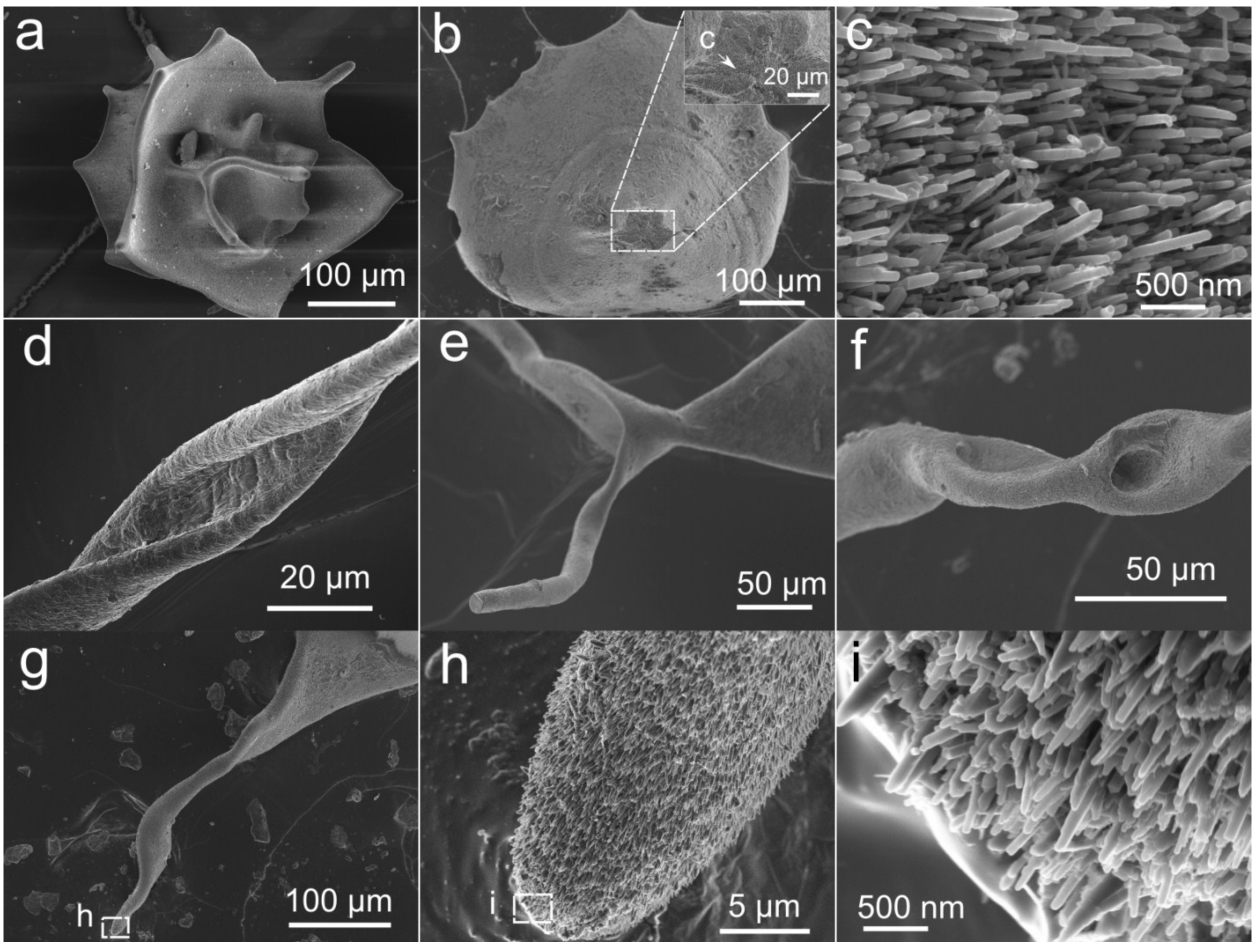

Fig. S5. FESEM micrographs of aragonite biomorphs formed at room temperature: (a) flower-like particle; (b) broken diabolo-like particle, and the inset shows the cross section of half of the dumbbell shaped core in the center; (c) close-up view of the co-oriented nanorods in the center of the cross section of the dumbbell shaped core; (d) helicoids developed from laminar sheet; (e) tentacle-like filament developed from laminar sheet; (f) tubular-like filament developed from laminar sheet; (g) the tentacle-like filament developed from laminar sheet at early stage; (h) close-up view of the growth front of the tentacle-like filament; (i) close-up view of the co-oriented nanorods in the growth front of the tentacle-like filament.

6. Time-lapse video of the growth of aragonite and calcite

Video S1. Time-lapse video of the growth of flower-like silica aragonite biomorphs and elongated calcite at room temperature.