

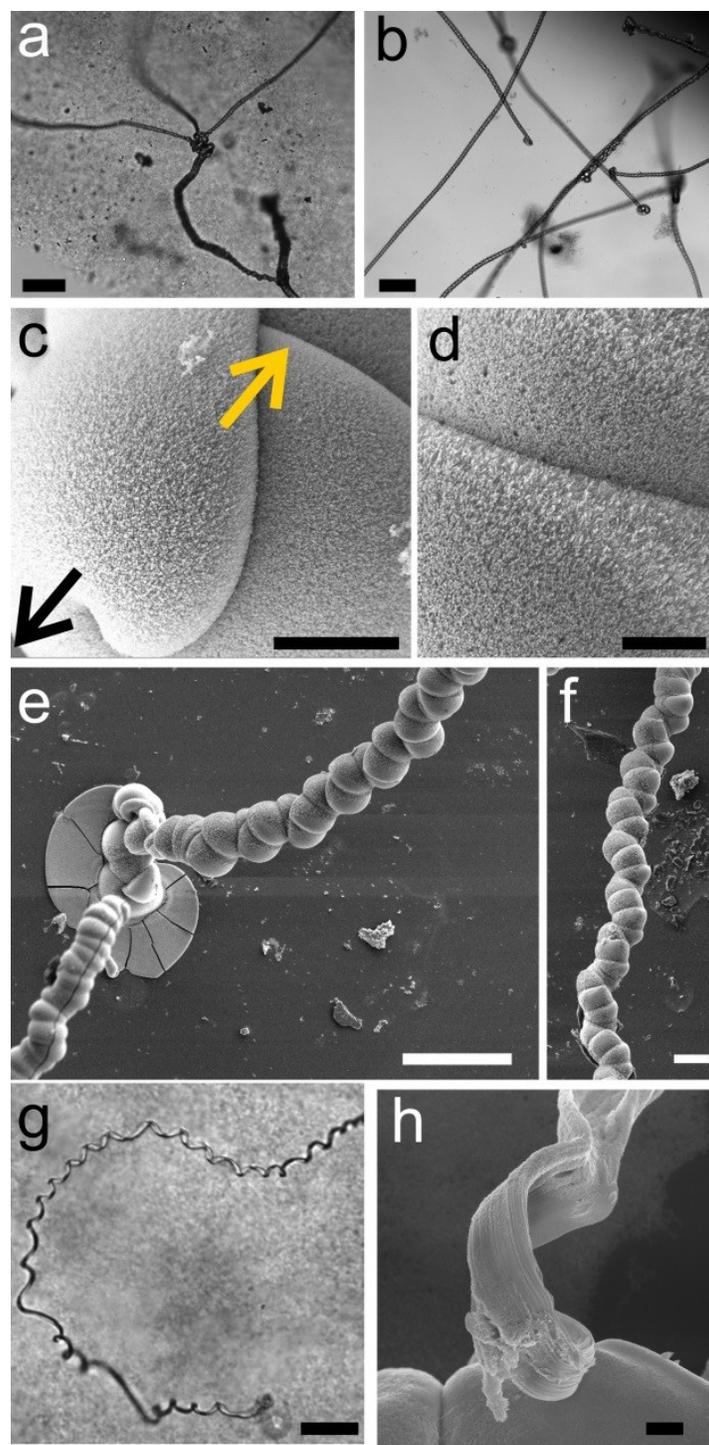
## ***Supplementary Information***

### **Systematic Characterization of Polycrystalline Silica-Carbonate Helices**

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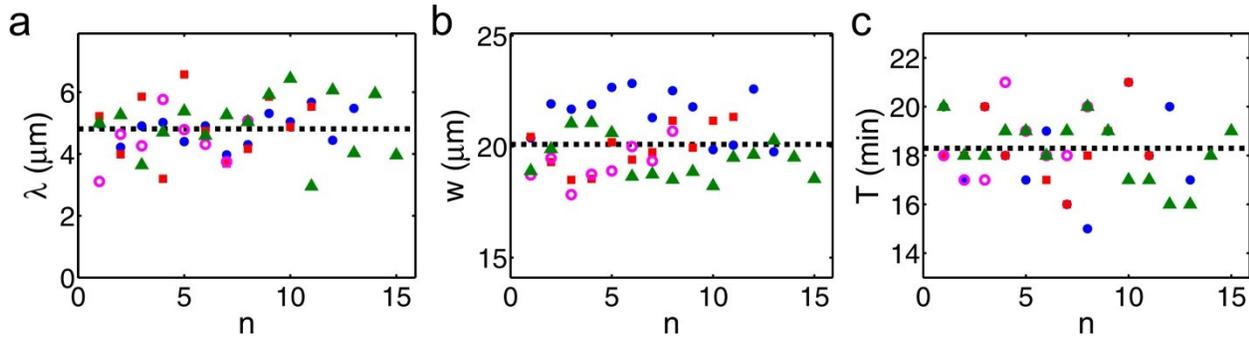
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We provide here additional micrographs of the different helical biomorphs. Figure S1a-b show networks of helices grown by the single-phase method that extend to millimeter lengthscales. These structures readily interconvert between the single and double helix growth modes. Figure S1c shows a side view of a single helix. The black arrow indicates the forward growth direction, while the yellow arrow represents the backwards overgrowth described in the text. Closer inspection of these structures reveals their distinctive layered morphology and the nanosized building blocks (Fig. S1d). Figures S1e-f and S1g-h show images of superhelices and ribbons, respectively.



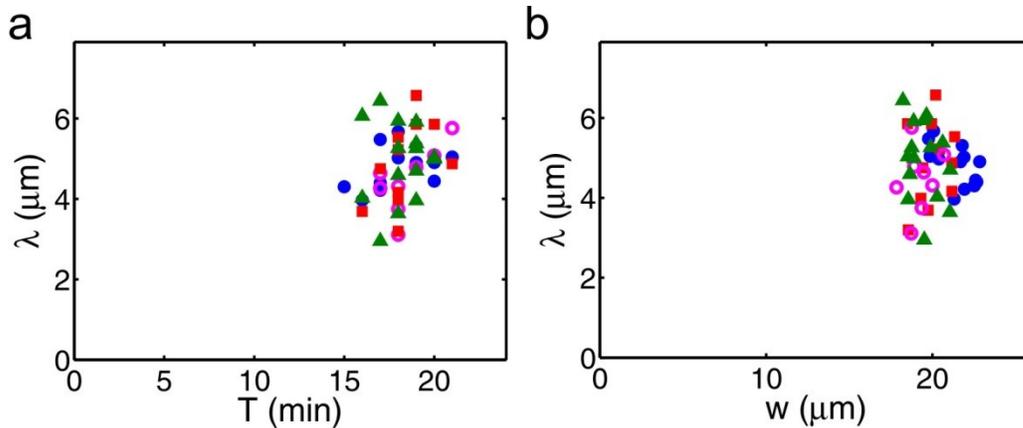
**Figure S1.** Optical and electron micrographs of (a,b) helix networks, (c,d) single helices, (e,f) superhelices, and (g,h) ribbons. The black and yellow arrows in (c) indicate the forward and backward growth directions, respectively. Scale bars: (a,b,g) 100  $\mu\text{m}$ , (e,f) 20  $\mu\text{m}$ , and (c,d,h) 3  $\mu\text{m}$ .

Detailed measurements of the single helix growth dynamics are provided in Fig. S2. The helix wavelength, width, and period fluctuate around the average values that are established almost immediately during helix growth (see manuscript text).



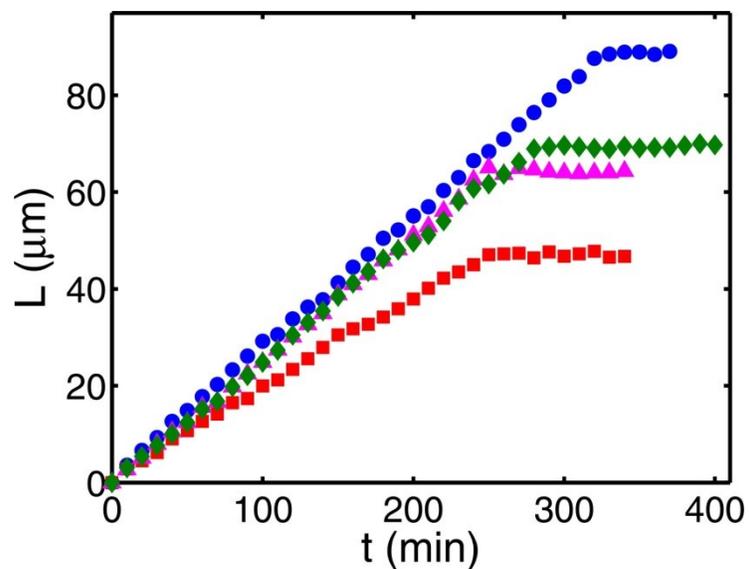
**Figure S2.** Evolution of single helix (a) wavelength, (b) width, and (c) period during growth.

The latter observation is verified by plotting the data in wavelength-period and the wavelength-width diagrams (Fig. S3). Recall that the double helices typically explore different variable pairs whose distribution is well described by linear functions (Fig. 5). In contrast, the single helices select characteristic mesoscopic values nearly instantaneously.



**Figure S3.** Distribution of the single helix wavelengths as a function of (a) growth period and (b) helix width.

Finally, we plot the single helix length as a function of time (Fig. S4). As discussed in the main manuscript text, our analyses show that the single helix extends at a constant velocity before abruptly ceasing its growth. This event is often accompanied by the nucleation of a new single or double helix at the side of the original structure.



**Figure S4.** Plot showing the single helix length as a function of growth time.