## Supporting Information for

## Structural Roles of Amphiphilic Peptide Tails on

## Silica Biomineralization

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**Materials.** The designed peptides were purchased from Shanghai Biotech Bioscience & Technology Co., Ltd, which have a purity of at least 95% (Figures S1 and S2). Tetraethyl orthosilicate (TEOS, 98%) and trimethoxysilylpropyl-N,N,N-trimethylammonium chloride (TMAPS, 50% in methanol) were purchased from Aldrich and Gelest, respectively. All chemicals were used as received without further purification.

**Sample Preparation.** The designed amphiphilic peptides were dissolved in pure water (18.2 M $\Omega$ •cm), and reached a weight concentration of 0.1%. Sodium hydroxide was added to adjust the pH to neutral (6.0–8.0) after the dissolution of the peptide. The designated amount of TMAPS was added to the homogeneous peptide solutions at room temperature. Then, TEOS was added to the mixed solution of the amphiphilic peptide and TMAPS. The oil drops of TEOS gradually disappeared, and the solution became cloudy and highly dense. The molar composition of the reaction mixture was peptide:TMAPS:TEOS = 1.0:4.0:16.0. One hour later, stirring was stopped, and the reaction mixture was aged at 353 K for 3 days. The powder product was recovered by centrifugation and freeze dried.

**Characterization.** SEM observation was performed with a field emission-scanning electron microscope JEOL JSM-7401F microscope at 0.8-1.0 kV. To observe the genuine external surface, the samples were observed without any metal coating. TEM observation was performed using a JEOL JEM-2100 microscope operated at 200 kV (Cs 1.0 mm, point resolution 0.23 nm). Images were recorded with a KeenView CCD camera (resolution 1376 x 1032 pixels, pixel size 6.45 x 6.45  $\mu$ m) at 30 000–120 000 times magnification under low-dose conditions. CD and UV/Vis spectra were recorded using a JASCO J-815 spectropolarimeter. The spectra baseline was corrected using pure water. The spectra were obtained at the peptide concentration of 0.01 mmol/L, and peptide:TMAPS:TEOS = 1.0:4.0:16.0. The spectra were recording by taking 3

averages from 190 to 350 nm with a 0.5-nm step and a 1-s collection time per step at room temperature. Thermogravimetric analyses were performed on Pyris 1 thermogravimetric analyzer (Perkin Elmer, Inc., USA). It was performed in air from 40°C to 750°C, and the heating rate was 20 °C/min.



Figure S1. HPLC analysis of P1-4 (a-d).



4.4E+004

2.2E+004

0.0E+000

456



592

728

342.3

864

1000

Figure S2. ESI-MS spectra of P1-4 (a-d).



**Figure S3.** Low magnification SEM images of nanoribbons prepared from P1 (a) and nanofibers prepared from P2 (b).



Figure S4. TGA curves of PTS1-3 (a-c).



Figure S5. TEM image of calcined PTS2, showing that the nanostructure was remained.