Supporting Information

Chemical Environment as Control Element in the Evolution of Shapes - ‘Hexagons and Rods’ from an 11-Helical $\alpha\beta^{2,3}$-hybrid Peptide

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<table>
<thead>
<tr>
<th>Table of Contents:</th>
<th>Page No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Representative large-area SEM images</td>
<td>3-4</td>
</tr>
<tr>
<td>2. TGA/DSC and powder XRD patterns of samples prepared from 1</td>
<td>5-6</td>
</tr>
<tr>
<td>3. $^1$H NMR and IR spectra of microcrystalline samples from 1</td>
<td>7</td>
</tr>
</tbody>
</table>
Larger area SEM images:

Figure S1. Larger area SEM images a-d) showing the time-dependent morphology changes in the samples of 1.

Figure S2. Larger area SEM images of samples of the tetrapeptide 1 formed by the addition of its 1 mg/mL solution in ACN (200 μL) to water containing various concentrations of P123: (a) 0.001, (b) 0.01, (c) 0.05, (d) 0.1, (e) 0.5, (f) 1.0, (g) 2.0, (h) 3.0, (i) 4.0, (j) 5.0 g/L.
**Figure S3.** Larger area SEM images showing different morphologies of samples of 1 formed by addition of its 1 mg/mL solution in THF (200 µL) to water containing various concentrations of P123: (a) 1.0, (b) 2.0, (c) 3.0, (d) 5.0 g/L.

**Figure S4.** Figures showing the morphology control on sequential use of organic co-solvents during the initiation and propagation phases of aggregation. a) 100 µL of a 2 mg/mL solution of 1 in ACN was added to 0.1 g/L P123-water (1 mL), and after stirring for 1 min, 100 µL of THF was added; b) 100 µL of a 2 mg/mL solution of 1 in THF was added to 0.1 g/L P123-water (1 mL) and after stirring for 1 min, 100 µL of ACN was added; c) 200 µL of a 1 mg/mL solution of 1 in 1:1 ACN-THF mixture was added to 0.1 g/L P123-water (1 mL).
**Figure S5.** Thermogravimetric analysis (TGA) of aggregates formed from 1 under various conditions.

**Figure S6.** a) DSC profile of the raw powder; b) Powder XRD patterns of the raw powder and single crystal (simulated from single crystal X-ray data).
Figure S7. Detailed DSC analysis of the sample (edge distorted hexagons) from peptide 1 prepared under ACN-0.01g/L P123-water, to see the reversibility of phase transitions. The experiment was conducted in two runs. The First heating was carried out from RT to 180°C which gave sharp endothermic peak at 169°C. The sample was then cooled to RT (First cooling). The absence of exothermic peak showed that the phase change at 169°C is irreversible. During the Second heating cycle we did not see the endothermic peak at 169°C as expected, but the peak at 195°C corresponding to melting point of the crystalline sample was present. Cooling of this sample (Second cooling) back to the RT did not involve any exothermic heat change at 195°C, indicating that the melting is accompanied by decomposition.

Figure S8. Development of hexagons from dendritic aggregates. Samples prepared under ACN-0.01g/LP123-water (Immediately after addition of ACN solution of peptide 1, an aliquot was withdrawn from the stirring mixture and analysed through SEM)
**Figure S9.** Comparison of the $^1$H NMR (400 MHz, CDCl$_3$) spectrum of authentic sample of 1 (a) with that of the samples prepared under 5g/L P123-THF (b), 1 g/L P123-ACN (c) and P123 alone (d); signals corresponding to traces of P123 can be seen in the spectra of microcrystalline aggregates.

**Figure S10.** Comparison of the FT-IR spectrum of the authentic sample of 1 with that of its microcrystalline aggregates.