Supporting Information

Crystallization and Self-assembly of Shape-Complementary Sequence-defined Peptoids

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Table S1. Characterization data of diblock copolypeptoids 1-6.

<table>
<thead>
<tr>
<th>Peptoid Nomenclature</th>
<th>Theoretical M</th>
<th>Observed[a]</th>
<th>Purity(%)b</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 Ac-DDNNOOHH-Nde8</td>
<td>2741.0</td>
<td>1371.5</td>
<td>914.7</td>
</tr>
<tr>
<td>2 Ac-HHOONNDD-Nde8</td>
<td>2741.0</td>
<td>1371.5</td>
<td>914.7</td>
</tr>
<tr>
<td>3 Ac-DHDHDDHDH-Nde8</td>
<td>2741.0</td>
<td>1371.5</td>
<td>914.7</td>
</tr>
<tr>
<td>4 Ac-HDHDDHDH-Nde8</td>
<td>2741.0</td>
<td>1371.5</td>
<td>914.7</td>
</tr>
<tr>
<td>5 Ac-DDHDDHHH-Nde8</td>
<td>2741.0</td>
<td>1371.5</td>
<td>914.7</td>
</tr>
<tr>
<td>6 Ac-HHDDHHDDH-Nde8</td>
<td>2741.0</td>
<td>1371.5</td>
<td>914.7</td>
</tr>
</tbody>
</table>

[a]Mass was obtained from LC-MS. [b]Purity was determined from LC-MS. N-decyl glycine is denoted as D and N-heptyl glycine is denoted as H.
**Figure S1.** LC-MS trace of purified DDNNOOHH-Nde₈ (1) with gradient of 50-95% acetonitrile in water in 6.8 min. Both acetonitrile and water contain 10% isopropanol. LC trace of 1 (top image) and mass spectrum of 1 (bottom image).

**Figure S2.** LC-MS trace of purified HHOONNDD-Nde₈ (2) with gradient of 50-95% acetonitrile in water in 6.8 min. Both acetonitrile and water contain 10% isopropanol. LC trace of 2 (top image) and mass spectrum of 2 (bottom image).
Figure S3. LC-MS trace of purified DHDHGDH-Nde₈ (3) with gradient of 50-95% acetonitrile in water in 6.8 min. Both acetonitrile and water contain 10% isopropanol. LC trace of 3 (top image) and mass spectrum of 3 (bottom image).

Figure S4. LC-MS trace of purified HDHDHGDH-Nde₈ (4) with gradient of 50-95% acetonitrile in water in 6.8 min. Both acetonitrile and water contain 10% isopropanol. LC trace of 4 (top image) and mass spectrum of 4 (bottom image).
Figure S5. LC-MS trace of purified DDHDDHHH-Nde$_8$ (5) with gradient of 50-95% acetonitrile in water in 6.8 min. Both acetonitrile and water contain 10% isopropanol. LC trace of 5 (top image) and mass spectrum of 5 (bottom image).

Figure S6. LC-MS trace of purified HHDDHDDH-Nde$_8$ (6) with gradient of 50-95% acetonitrile in water in 6.8 min. Both acetonitrile and water contain 10% isopropanol. LC trace of 6 (top image) and mass spectrum of 6 (bottom image).
Figure S7. MALDI-TOF spectrum of Ac-DDNNOOH-Nde$_8$ (1).
Figure S8. MALDI-TOF spectrum of Ac-HHOONDD-Nde₈ (2).
Figure S9. MALDI-TOF spectrum of Ac-DHDHDHDH-Nde₈ (3).
Figure S10. MALDI-TOF spectrum of Ac-HDHDHDHD-Nde₈ (4).
Figure S11. MALDI-TOF spectrum of Ac-DDHDDHH-Nde₈ (5).
Figure S12. MALDI-TOF spectrum of Ac-HHDDHHDD-Nde₈ (6).
Figure S13. Chemical structures and DSC traces of peptoids with non-complementary molecular shapes: (A) Ac-HHDDDDHH-Nde₈ and (B) Ac-D₉D₉PPPD₉D₉-Nde₈. N-decyl glycine is denoted as D, N-heptyl glycine is denoted as H, N-pentyl glycine is denoted as P, N-dodecyl glycine is denoted as D₉.
Figure S14. TEM images of sheet 2-6 (A-E).
Figure S15. NanoDSC traces of diluted nanosheets in milliQ water at 2 mg/mL.
Figure S16. Diffraction patterns of sheet 3 and 4.