

## **Supporting Information**

*of*

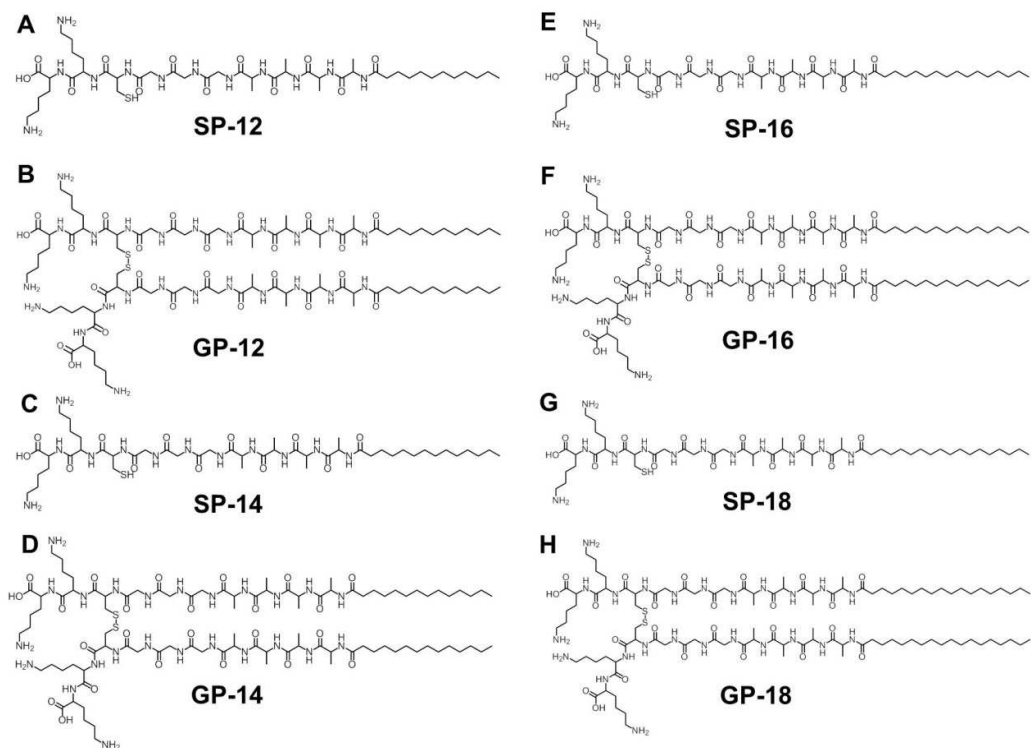
### **Controlled Arrays of Self-assembled Peptide Nanostructures in Solution and at Interface**

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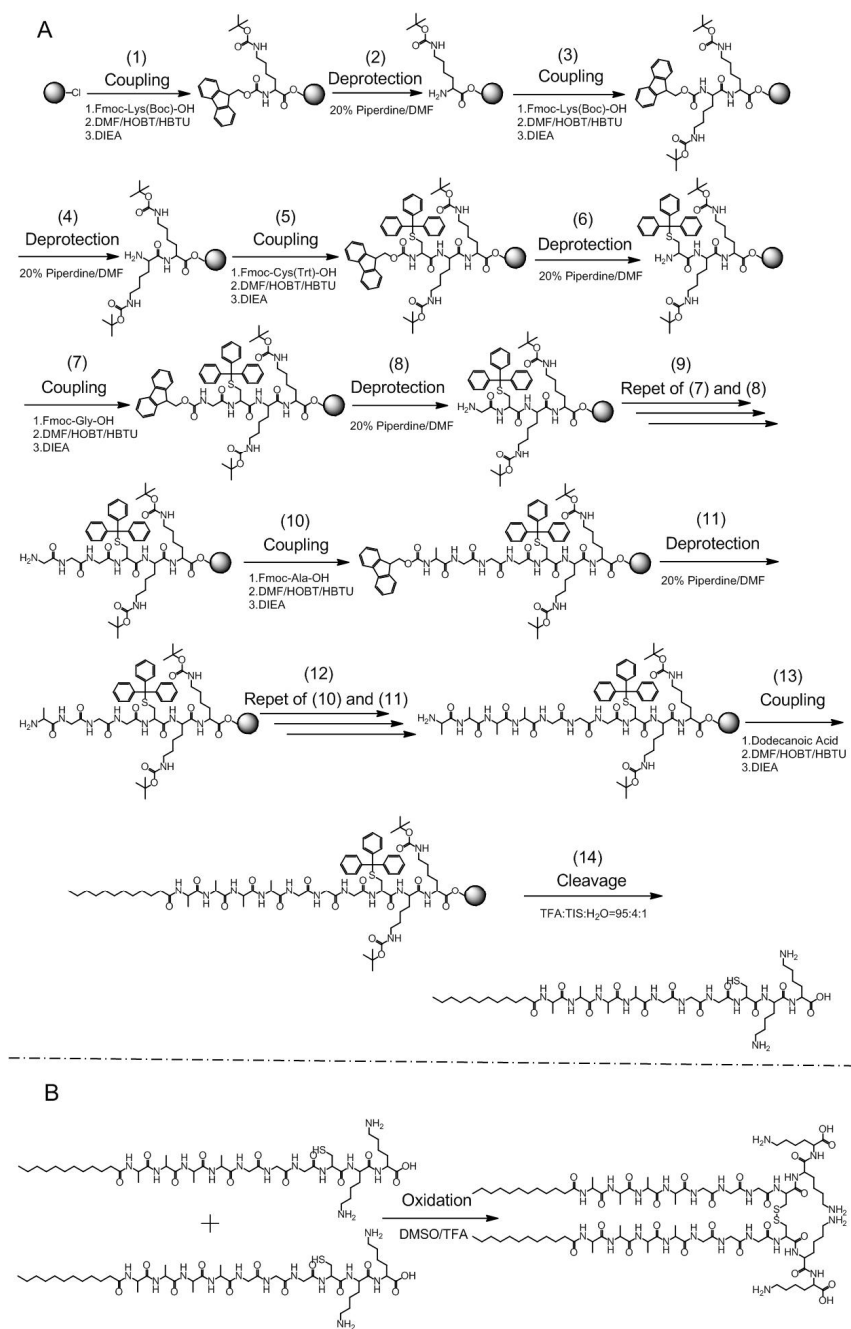
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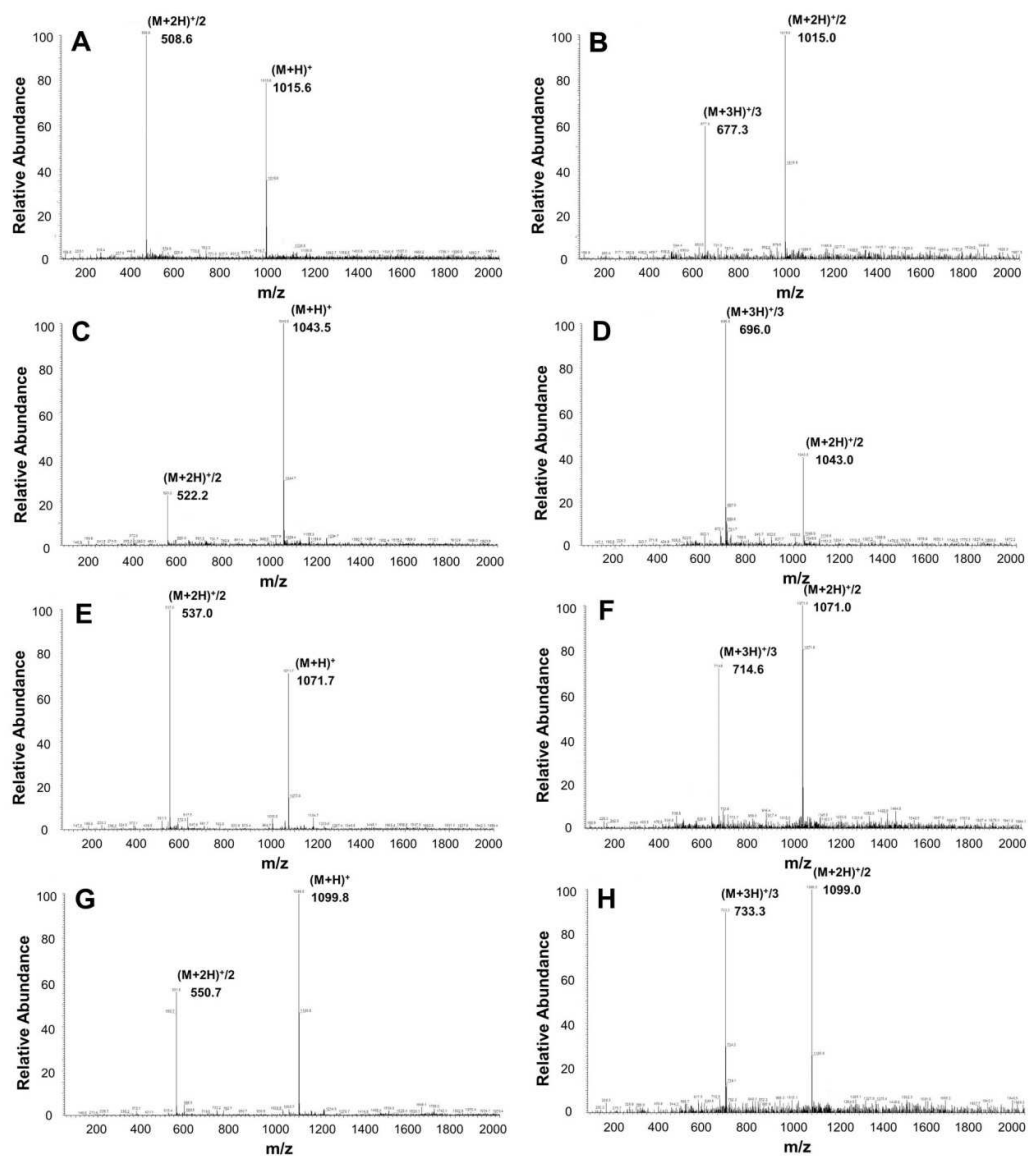
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**Scheme S1.** The molecular structures of the SAPs and GAPs. (A) SAP-12, (B) GAP-12, (C) SAP-14, (D) GAP-14, (E) SAP-16, (F) GAP-16, (G) SAP-18, and (H) GAP-18.



**Scheme S2.** Synthesis of (A) SAP-12 and (B) GAP-12.



**Figure S1.** The ESI-MS profiles of the SAPs and GAPs: (A) SAP-12, (B) GAP-12, (C) SAP-14, (D) GAP-14, (E) SAP-16, (F) GAP-16, (G) SAP-18, and (H) GAP-18.

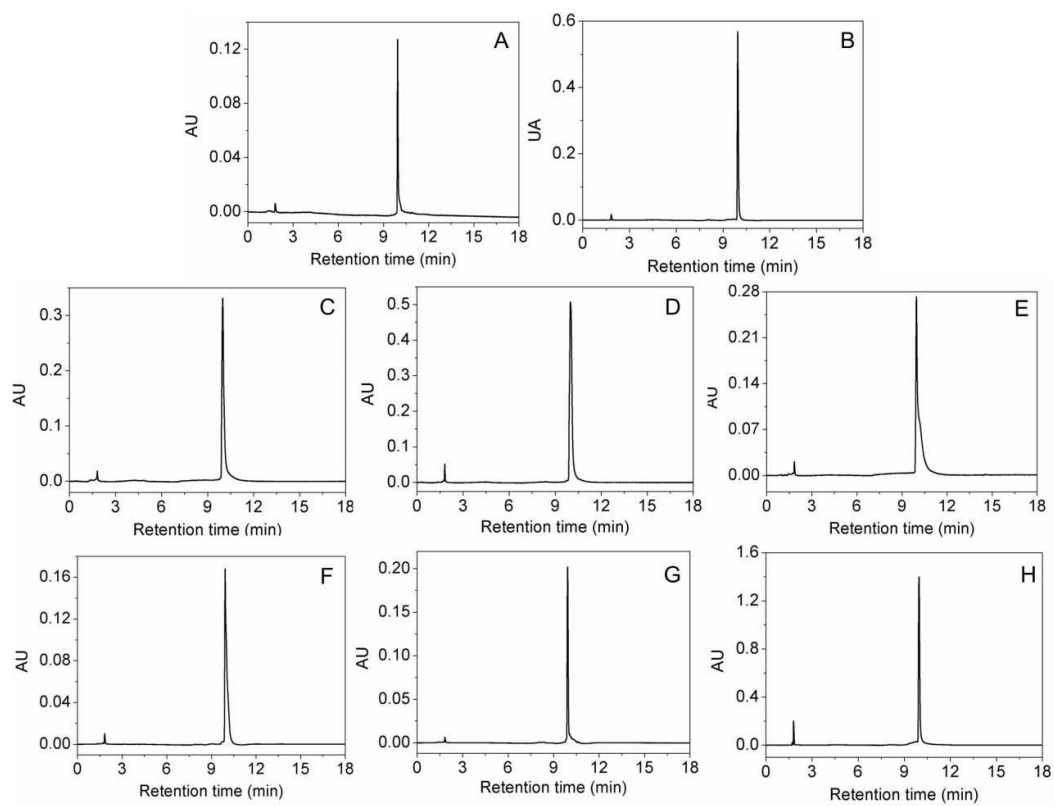
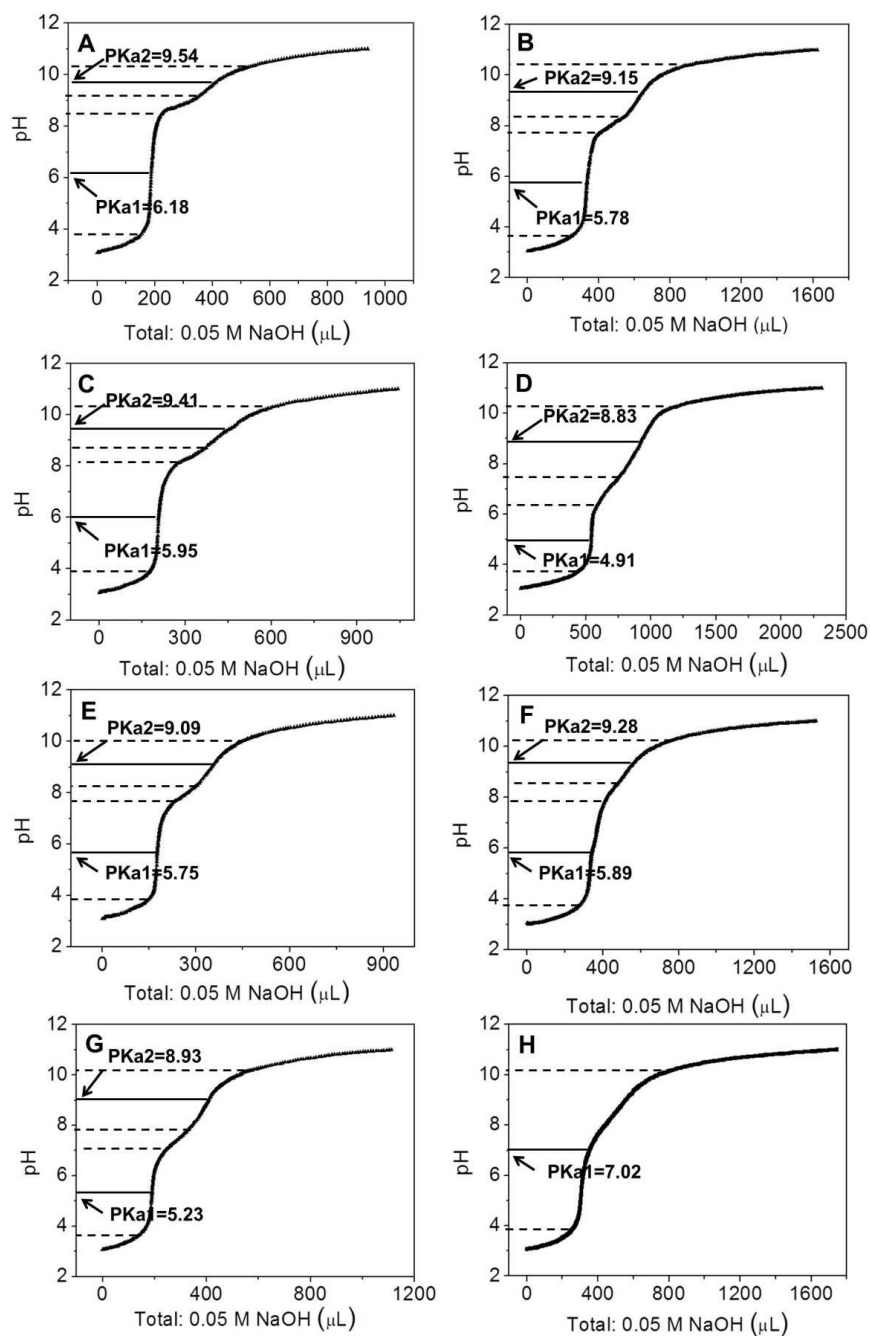
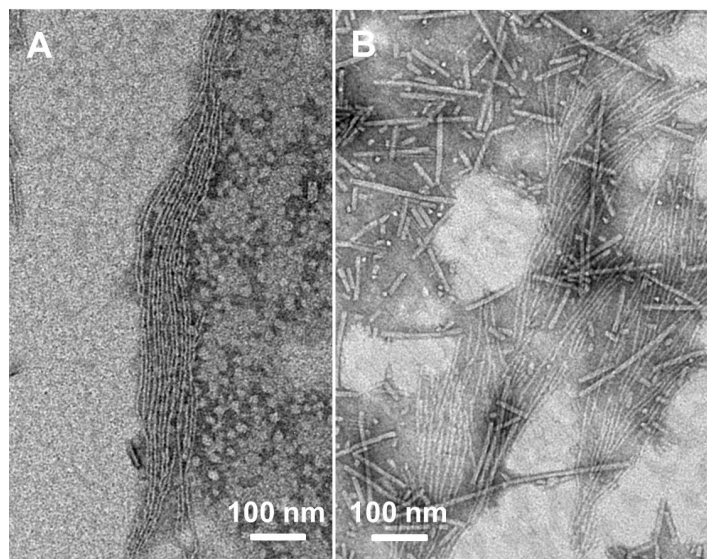


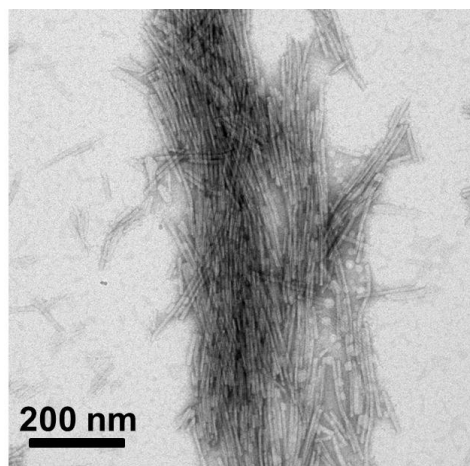
Figure S2. HPLC analysis of the SAPs and GAPs: (A) SAP-12, (B) GAP-12, (C) SAP-14, (D) GAP-14, (E) SAP-16, (F) GAP-16, (G) SAP-18, and (H) GAP-18.



**Figure S3.** Acid-base titration curves of the peptide building blocks: (A) SAP-12, (B) GAP-12, (C) SAP-14, (D) GAP-14, (E) SAP-16, (F) GAP-16, (G) SAP-18, and (H) GAP-18.

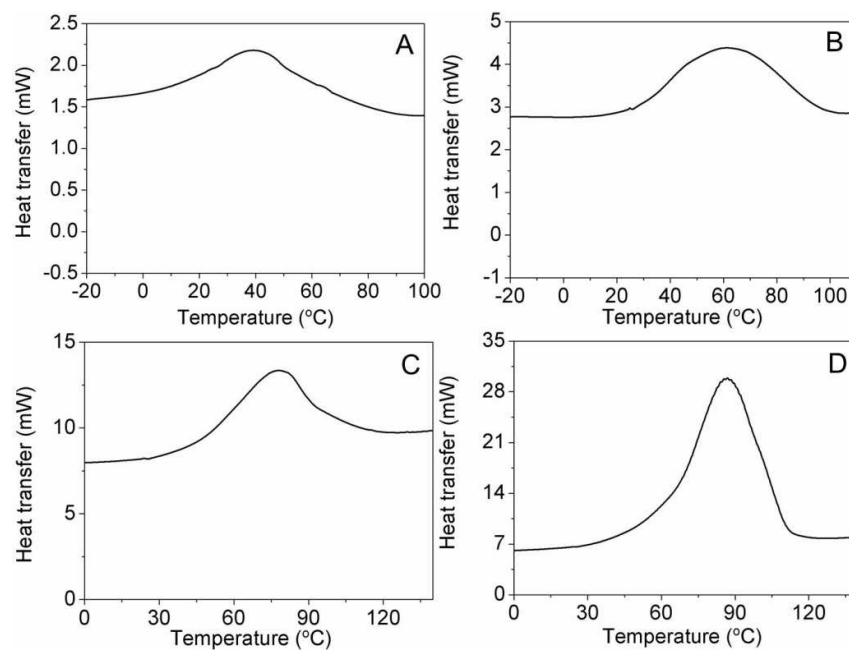


**Figure S4.** TEM images of fibers formed by SAP-12 (A) and GAP-12/SAP-12 1/3  
(B).



**Figure S5.** TEM images of hierarchical assembly of short rod-like fibers formed by the co-assembly of SAP-12 and GAP-12 into a fibrous alignment in the same direction at 30 °C for 24 h.





**Figure S6.** DSC analysis of the GAPs: (A) GAP-12, (B) GAP-14, (C) GAP-16, and (D) GAP-18.

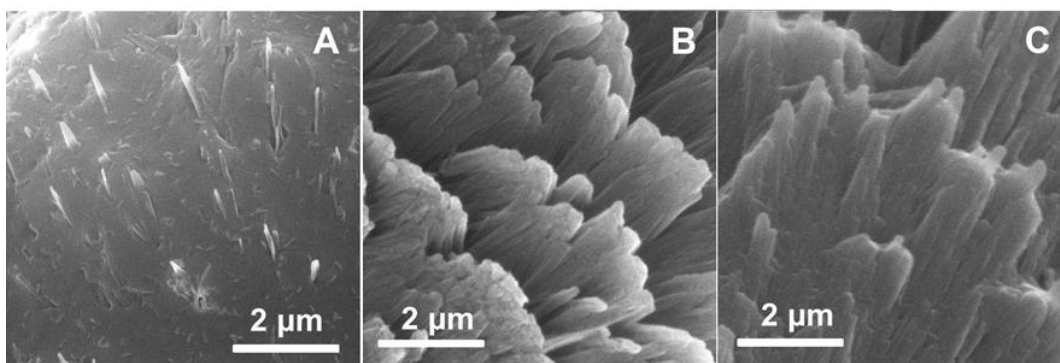


Figure S7. SEM images of vertically aligned array of peptide nanostructures formed by SAP-12/GAP-12 on the hydrophilic naked silica wafer at the different concentrations: 0.01 mg/ml (A), 10 mg/ml (B) and 20 mg/ml (C).

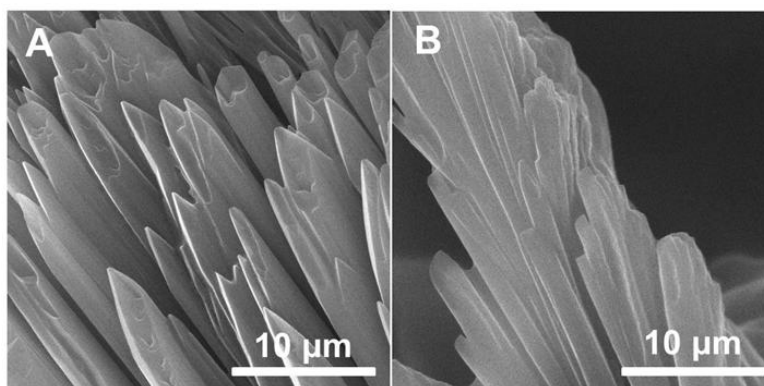


Figure S8. SEM images of the interfacial assemblie of SAP-12/GAP-12 on the hydrophobic interface with a contact angel  $91.7^{\circ}$  at the different concentrations: 15 mg/ml (A), and above 20 mg/ml (B).