Supporting Information

Interfacial Adsorption of Cationic Peptide Amphiphiles: a Combined Study of \textit{in situ} Spectroscopic Ellipsometry and Liquid AFM

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Figure S1. HPLC profiles of (a) G6K, (b) A6K, and (c) V6K. The conditions for HPLC analysis of the three peptides are as follows: eluent A, 0.1% trifluoroacetic acid in water, 0→1min, 95% (A%), 2→20min, 95→40% (A%); eluent B, 0.1% trifluoroacetic acid in acetonitrile; UV, 214nm; flow rate, 0.8mL/min; column, RP-18, 4.6mm×150mm. The measurements were performed on a Waters 2695 Alliance HPLC system at the temperature of 25 °C.
Figure S2. MALDI-TOF mass spectra of (a) G₆K, (b) A₆K, and (c) V₆K. The measurements were performed on Bruker Biflex III matrix assisted laser desorption/ionization time of flight (MALDI-TOF) mass spectrometer equipped with a 337 nm nitrogen laser, and 4-hydroxy-α-cyanocinnamic acid was used as the matrix. The samples were dissolved with the matrix in 1:1 (v/v) acetonitrile : water with 1% trifluoroacetic acid. Around 0.5 μl of the sample solution was placed on a metal sample plate and then allowed to air-dry at ambient temperature. Mass spectra were
acquired in positive linear mode and using an acceleration voltage of 19 kV. External mass calibration was performed using a standard peptide mixture. Spectra were obtained by setting the laser power close to the threshold of ionization and generally 100 pulses were acquired and averaged.

The calculated molecular masses for the three peptides are all well consistent with the observed as follows:


It is important to note that besides the distinguished singly charged molecular ion peaks, no other peak as well as fragmental ion peak was observed, indicating the high purity of our peptide samples.
Figure S3

**Figure S3.** TEM micrograph of peptide nanofibers formed in the 2.0 mM AβK solution at pH 6.0. The sample was negatively stained with uranyl acetate.
Figure S4

*Figure S4.* CD spectra measured from a) 2.0 mM A$_6$K and b) 0.2 mM V$_6$K solutions at pH 6.0 and the ambient temperature of 23 °C, indicating the formation of β-sheet conformation in both cases.
Figure S5

Figure S5. Typical height sectional profile of V$_6$K fibrillar aggregates formed at the concentration of 0.05 mM, giving heights of 3-4 nm.
Figure S6

Figure S6. Typical height sectional profile of V₆K globular stacks at the concentration of 0.2 mM, giving heights of 6-10 nm and diameters of 150-300 nm.
Figure S7

*Figure S7.* a) In situ height AFM image (10 μm×10 μm) of A6K at the silica/water interface at pH 6.0, with the peptide concentration fixed at 2.0 mM and b) the corresponding in situ phase image (10 μm×10 μm). The Z scale is shown to the right of the images, being A) 30 nm and a) 20 °, respectively.
**Figure S8**

**Figure S8.** Typical height sectional profile of A6K surface aggregates at 2.0 mM, giving heights of 5-10 nm.