Electronic Supplementary Information

Selective Disruption of Each Part of Janus Molecular Assemblies by Lateral Diffusion of Stimuli-Responsive Amphiphilic Peptide

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1. Amphiphilic polypeptides, **SHLL**, **SLL** and **SDL**

pH-responsive amphiphilic polypeptide **SHLL** used in this paper was the same compound used in previous report.[1] The other amphiphilic polypeptides, **SLL** and **SDL** also are the same compounds in our reports.[2, 3] The identification and synthesis of them were shown there.[1-3]

2. pK_a value of the His residues in **SHLL** aggregate

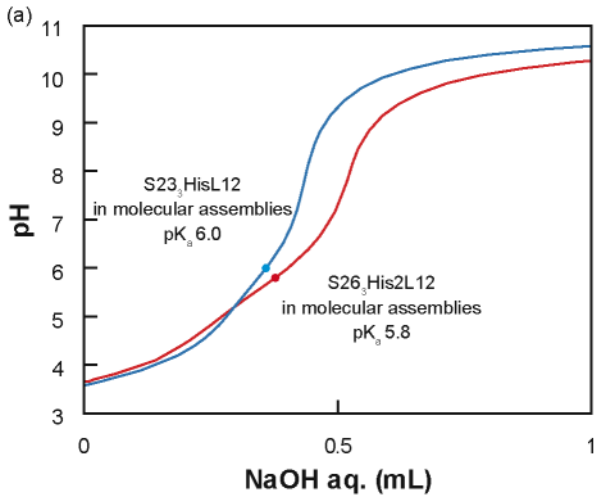

Fig. S1. The experimental titration curves obtained for aqueous solutions of aggregate prepared from ((Sar)$_{26}$)$_3$-*b*-((L-His)$_2$-(L-Leu-Aib)$_6$) (**SHLL**) (red line). This data has already been shown in previous report.[1]

3. Wide-area TEM images

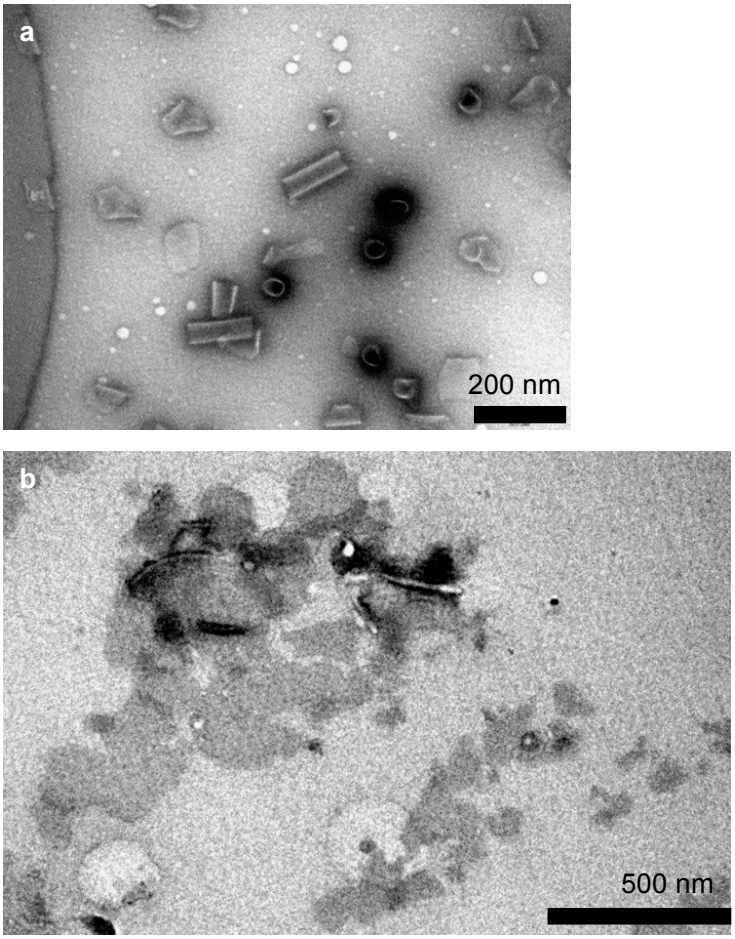

Fig. S2. TEM images of molecular assemblies composed of a single component of **SHLL** (a) and an equimolar mixture of **SHLL** + **SDL** (b) after changing pH from 7.4 to 5.0 and heating at 70 °C for 1 h.

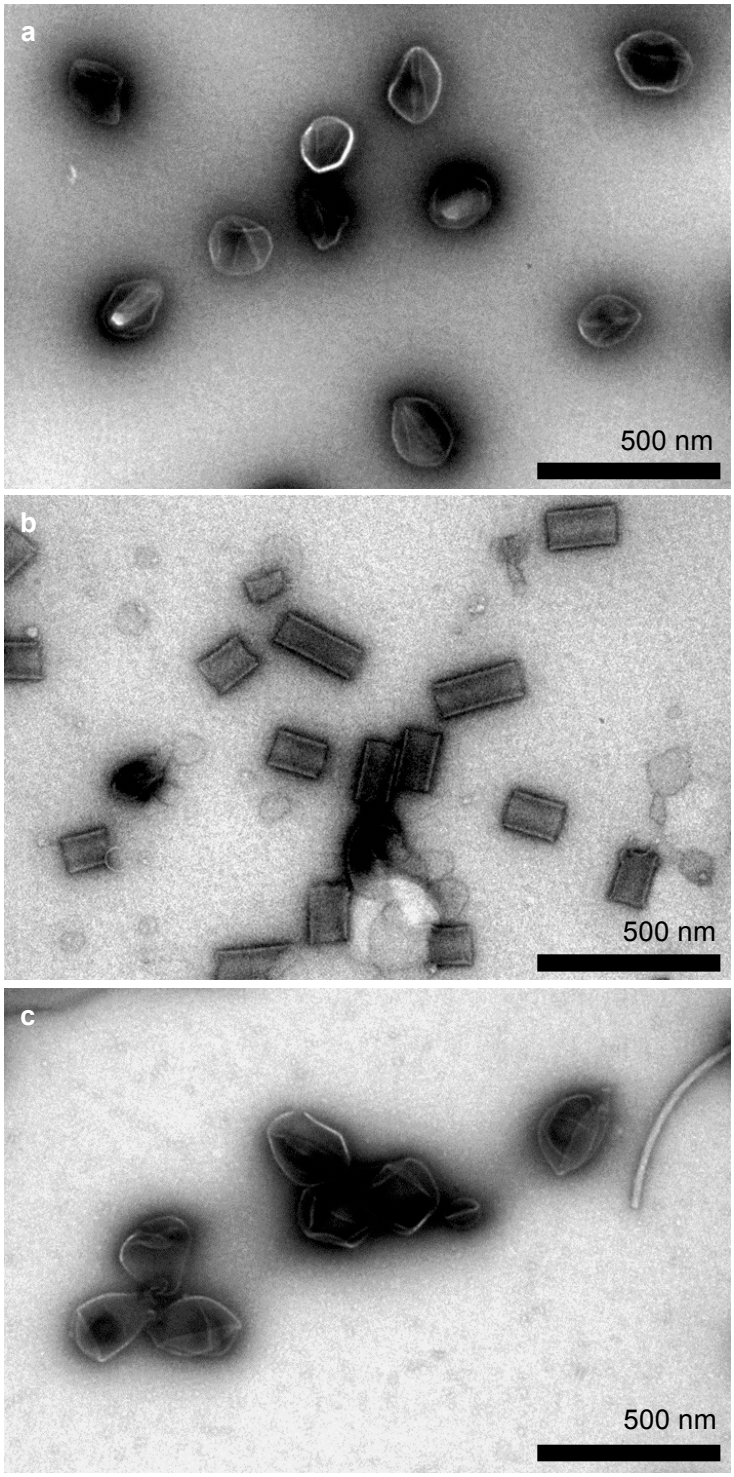

Fig. S3. TEM images of molecular assemblies prepared from a mixture of **SHLL** nanotube and **SLL** + **SDL** vesicle (a), **SDL** nanotube and **SHLL** + **SDL** vesicle (b) and **SLL** nanotube and **SHLL** + **SDL** vesicle (c) after changing pH from 7.4 to 4.7 (a) or 5.0 (b, c) and heating at 70 °C for 1 h.

4. Stability of **SLL** + **SDL** vesicle at pH 5 with the heat treatment

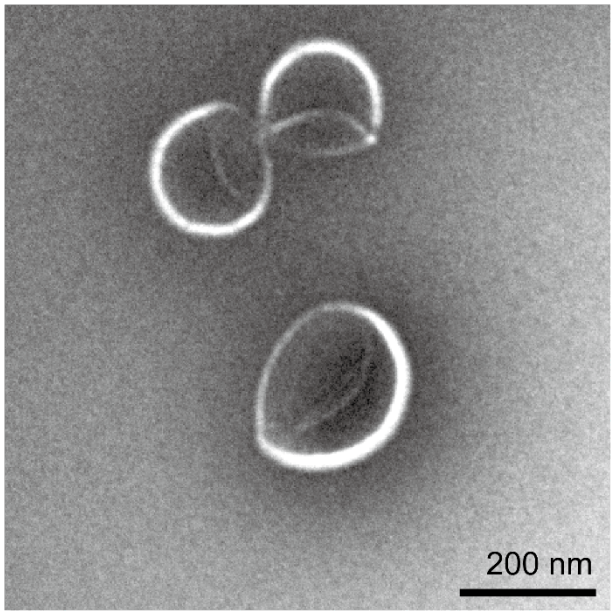

Fig. S4. TEM images of **SLL** + **SDL** vesicle at pH 5 with the heat treatment (70 °C, 1 h).

5. Diameter of the neck part in three kinds of round-bottom flask-shaped assemblies

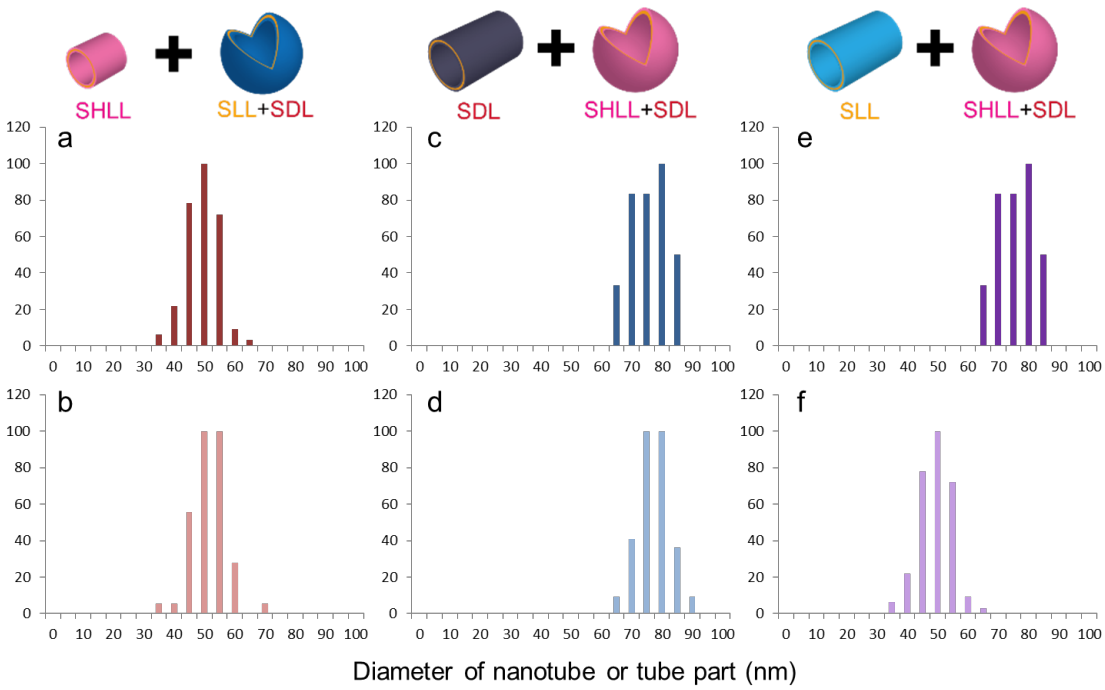

Fig. S5. The diameter of the nanotube before heating (a, c, e) and the tube part in the round-bottom flask-shaped assembly after heating (b, d, f) about three kinds of patchwork assemblies prepared from the **SHLL** nanotube and the **SLL** + **SDL** sheets (a, b), the **SDL** nanotube and the **SHLL** + **SDL** sheets (c, d), and the **SLL** nanotube and **SHLL** + **SDL** sheet (e, f). All assemblies were heated at 90 °C for 1 h.

Reference

1. Uesaka A, Ueda M, Makino A, Imai T, Sugiyama J, Kimura S. Morphology Control between Twisted Ribbon, Helical Ribbon, and Nanotube Self-Assemblies with His-Containing Helical Peptides in Response to pH Change. Langmuir. 2014;30(4):1022-1028.

2. Ueda M, Makino A, Imai T, Sugiyama J, Kimura S. Transformation of peptide nanotubes into a vesicle via fusion driven by stereo-complex formation. Chem Commun. 2011;47(11):3204-3206.

3. Kanzaki T, Horikawa Y, Makino A, Sugiyama J, Kimura S. Nanotube and Three-Way Nanotube Formation with Nonionic Amphiphilic Block Peptides. Macromol Biosci. 2008;8(11):1026-1033.