**Supplementary Information**

**Interior-filled Self-assemblies of Tyrosyl Bolaamphiphiles Regulated by Hydrogen Bonds**

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1. TEM image of Tyr-C7 assemblies

Tyr-C7 assembly was investigated using TEM at high magnification. Microdomains could not be observed because the hydrophobic and hydrophilic segments are too small to create visible, discrete domains. The zoomed image shows some domain-like pattern, however, it is not distinguishable from such patterns that observed in background.

Figure S1. TEM images of Tyr-C7 assembly at (a) low and (b) high magnification.
2. Formation of small Tyr-C7 assemblies by addition of GuHCl

When GuHCl was added to the pre-formed Tyr-C7 assembly in ~200 nm, the assembly was dissociated to produce tiny particles.

![SEM image of small Tyr-C7 aggregates](image)

**Figure S2.** SEM image of small Tyr-C7 aggregates (d = ~10 nm) produced by addition of GuHCl to the pre-formed Tyr-C7 assemblies (d = ~200 nm).
3. Dissolution of Tyr-C7 in 1,4-butanediol and ethanol

Tyr-C7 powder did not dissolve in 1,4-butanediol and ethanol, and remained crystallized powders. Based on these results, Tyr-C7 was turned out to dissolve and to form spherical assembled structures in polar solvents with dielectric constants higher than ~ 30.

Figure S3. SEM images of crystallized Tyr-C7 powders remaining in (a) 1,4-butanediol and (b) ethanol.