Supporting Information

Crosslinking Induced Self-organization of Polymers into Degradable Assemblies

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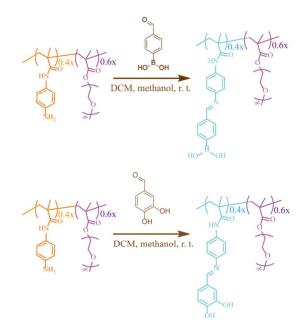
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Scheme S1. Synthetic procedures of BP and CP.

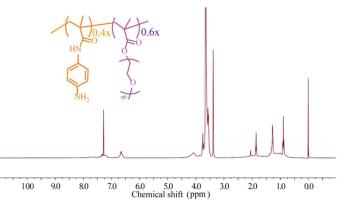


Figure S1. ¹H NMR spectrum of P(APMA-co-MAPEG) in CDCl₃.

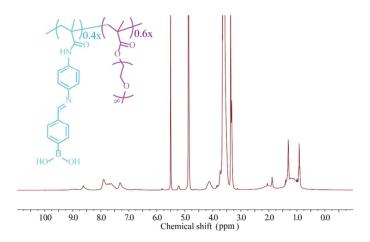


Figure S2. ¹H NMR spectrum of BP in methanol-D₄.

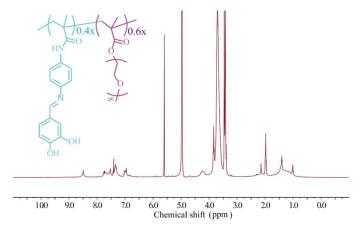


Figure S3. ¹H NMR spectrum of CP in methanol-D₄.

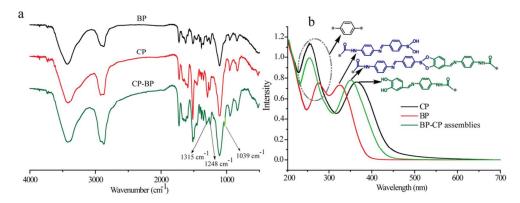


Figure S4. FT-IR (a) and UV-vis spectra (b) of BP, CP and BP-CP assemblies. The absorption peaks were tentatively assigned to the corresponding groups as shown in (b).

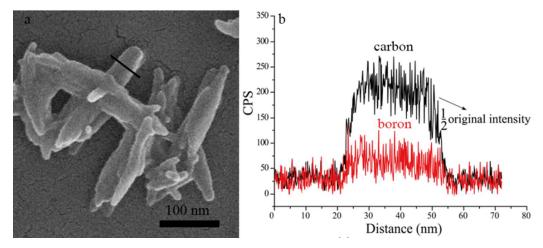


Figure S5. SEM image of bundle-like assemblies redispersed in water solution (a) and the corresponding EDX line scan analyses (b).

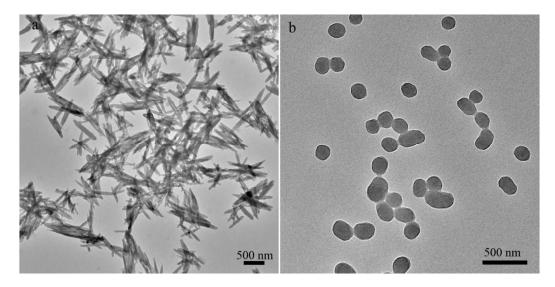


Figure S6. TEM images of bundle-like assemblies (a) formed in dichloromethane with a concentration of BP+CP 2.0 mg/mL, and spherical assemblies (b) formed in a mixed solvent (the volume ratio of methanol to water is 1:1) with 2.0 mg/mL BP+CP.

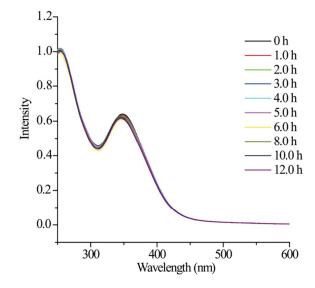


Figure S7. UV-vis spectrum evolution of bundle-like assemblies at pH=7.4 without D-glucose. Bundle-like assemblies were prepared by using 0.2 mg/mL of BP and CP.

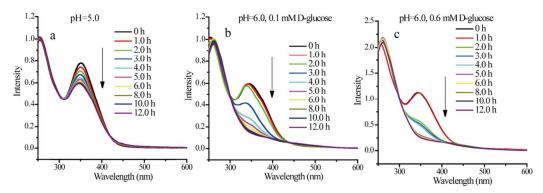


Figure S8. UV-vis spectrum evolution of bundle-like assemblies under different stimuli: (a) pH=5.0, (b) pH=6.0 with 0.1 mM of D-glucose, (c) pH=6.0 with 0.6 mM of D-glucose. Bundle-like assemblies were prepared by using 0.2 mg/mL of BP and CP.

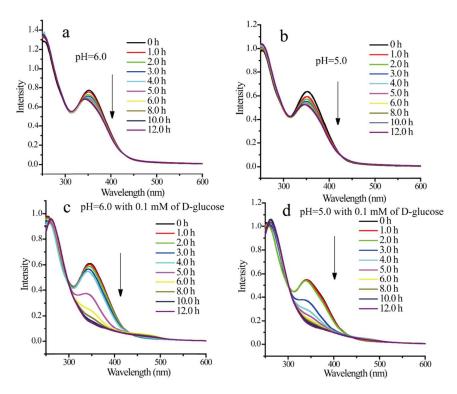


Figure S9. UV-vis spectrum evolution of spherical assemblies under different stimuli: (a) pH=6.0, (b) pH=5.0, (c) pH=6.0 with 0.1 mM of D-glucose and (d) pH=5.0 with 0.6 mM of D-glucose. Spherical assemblies were prepared by using 0.2 mg/mL of BP and CP.

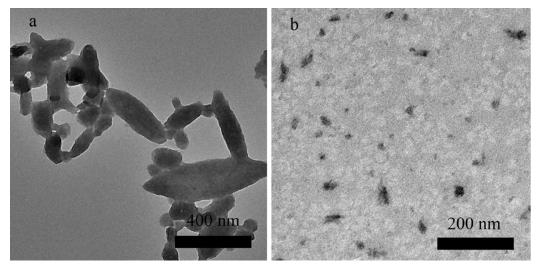


Figure S10. TEM images of bundle-like assemblies after staying in a solution with 0.1 mM of D-glucose at pH=5.0 for 2.0 h (a) and 8.0 h (b). Bundle-like assemblies were prepared by using 0.2 mg/mL of BP and CP.

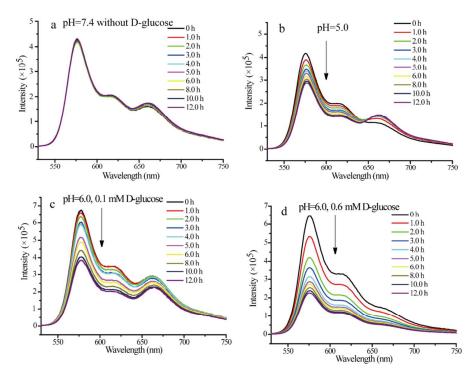


Figure S11. Fluorescence spectrum evolution of DiI encapsulated in bundle-like assemblies in a solution at pH=7.4 without D-glucose (a). Fluorescence spectra trace the release of DiI from bundle-like assemblies in response to different stimuli: (b) pH=5.0, (c) pH=6.0 with 0.1 mM of D-glucose, (d) pH=6.0 with 0.6 mM of D-glucose. Bundle-like assemblies were prepared by using 0.2 mg/mL of BP and CP.

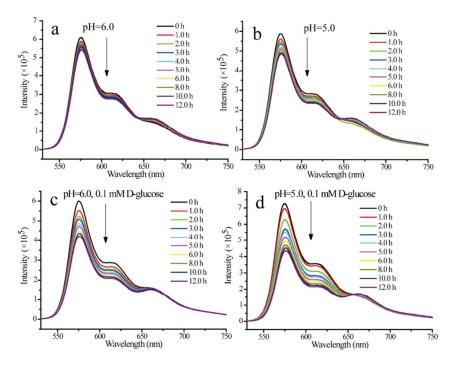


Figure S12. Fluorescence spectra trace the release of DiI from spherical assemblies in response to different stimuli: (a) pH=6.0, (b) pH=5.0, (c) pH=6.0 with 0.1 mM of D-glucose, (d) pH=5.0 with 0.1 mM of D-glucose. Spherical assemblies were prepared by using 0.2 mg/mL of BP and CP.

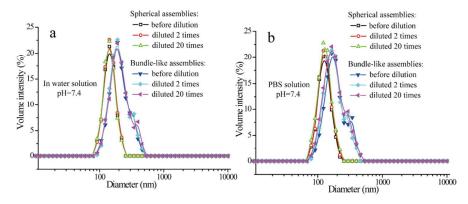


Figure S13. Stability of spherical and bundle-like assemblies against dilution: (a) in water solution with pH=7.4, (b) in PBS solution with pH=7.4. Both spherical and bundle assemblies were prepared by using 0.2 mg/mL BP+CP. Therefore, in this measurement, samples before dilution, diluted 2 times and diluted 20 times represented assembly concentration of 0.2, 0.1 and 0.01 mg/mL, respectively. The DLS tests were conducted 24 h after dilution.

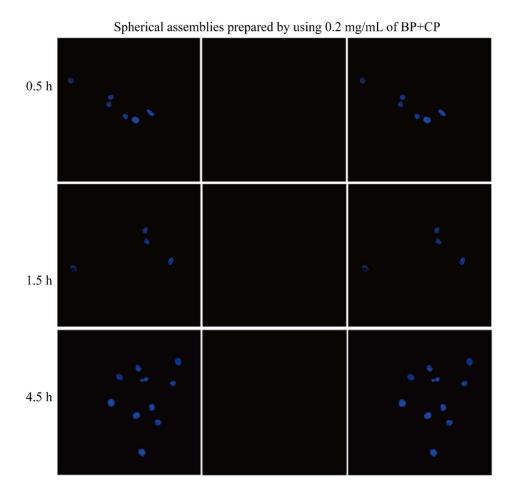


Figure S14. CLSM images of Hela cells incubated with spherical assemblies prepared by using 0.2 mg/mL of BP and CP. For each panel, the images from left to right show cell nuclei stained with 2-(4-amidinophenyl)-6-indolecarbamidine dihydrochloride (DAPI, blue), DiI loaded BP-CP assemblies (red) in cells, and overlays of the two images.