

Supporting Information

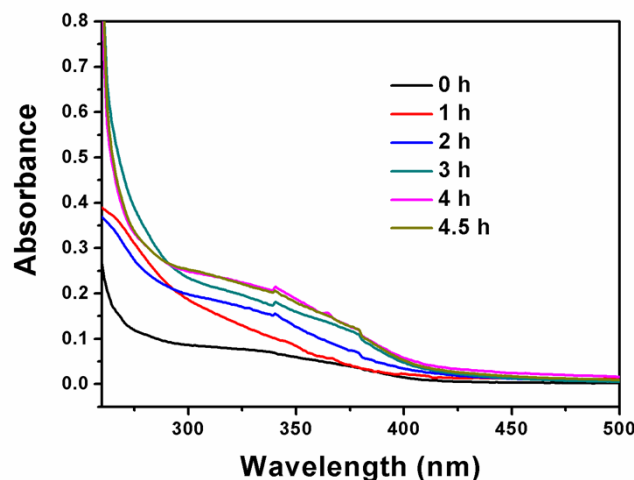


Figure S1. UV-vis spectra of 0.5 mg/mL L PNBC₁₀ in mixed solvents (DMSO/acetonitrile = 6/4, v/v) after 365 nm irradiation of different times. The photolysis ratio can also be calculated by 1 H NMR and/or from the change of the absorbance at 310 nm at different times according to our previous publications.^{1,2}

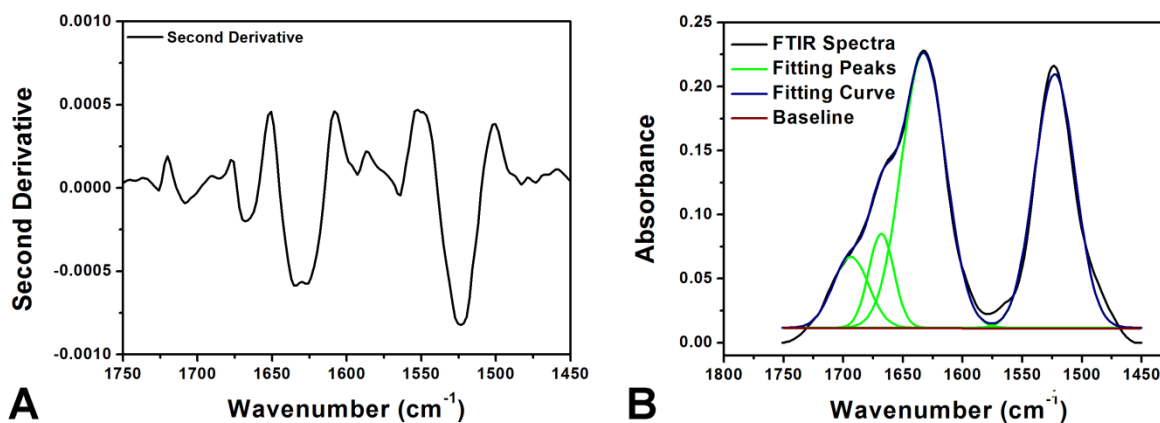


Figure S2. (A) Second derivative and (B) FT-IR spectra and the related fitting curve and fitting peaks for L PNBC₁₀ as an example.

Table S1. Secondary conformations of the graft polypeptides 1, 2, and 3 in solid state.

Entry	β -sheet (%)	Others (%)
^L PNBC ₁₀	83.6	16.4
1a	44.6	55.4
1b	32.9	67.1
1c	25.4	74.6
2a	37.5	62.5
2b	14.6	85.4
3a	34.0	66.0
3b	19.4	80.6

Table S2. The secondary conformations of 1, 2, and 3 in aqueous solution.

Entry	α -helix (%)	β -sheet (%)	Trn ^a (%)	Unrd ^b (%)
1a	16.0	36.5	17.1	30.4
1b	21.8	28.5	24.6	25.1
1c	40.7	9.1	26.8	23.4
2a	4.0	40.9	23.0	32.2
2b	16.7	24.0	25.7	33.6
3a	10.8	38.6	26.5	24.1
3b	17.1	25.2	26.8	30.9

a: Trn denotes turn; b: Unrd denotes unordered structures; all these data were calculated by using the CDPRO software according to a continll method.

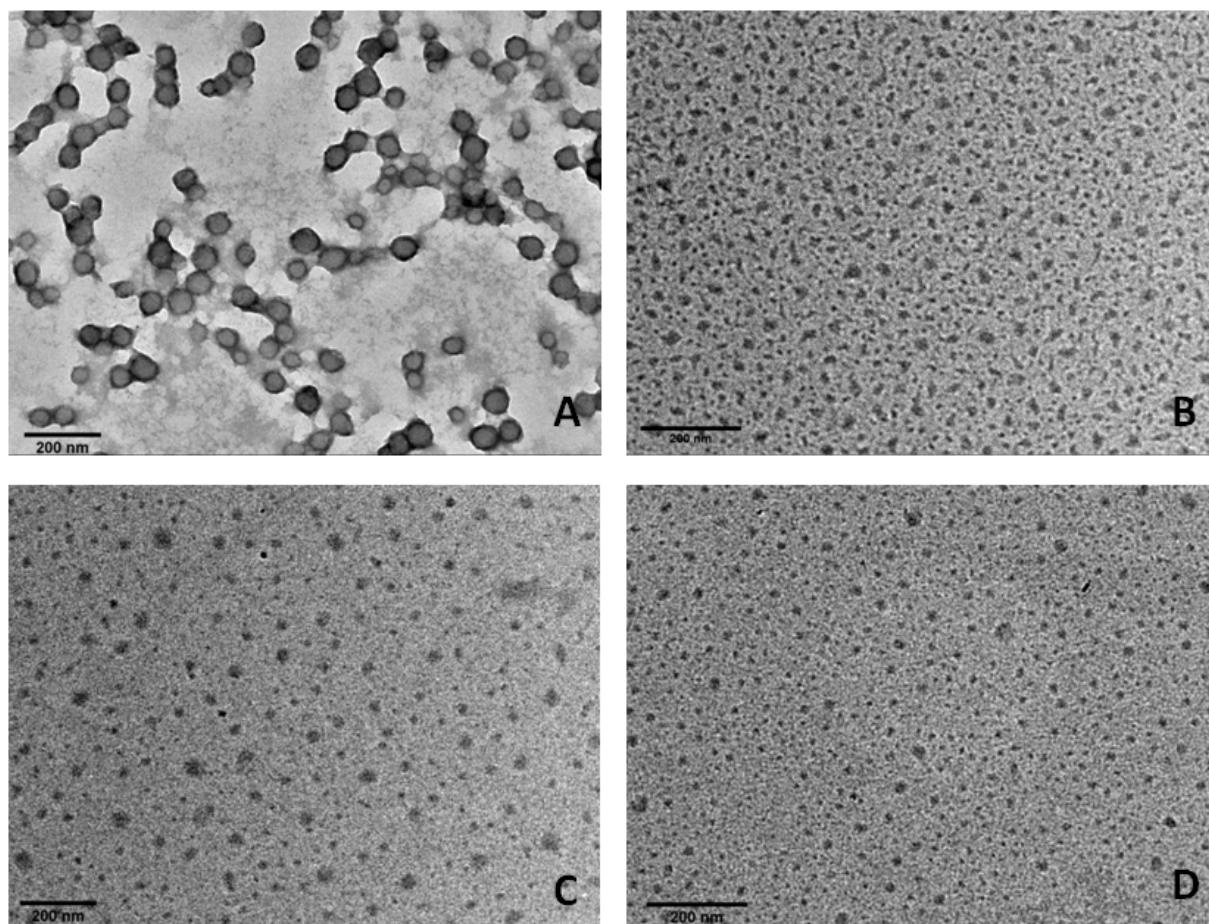


Figure S3. TEM images of 2a (A) and the pH (pH = 12) induced micelles from 2a (B); the self-assembled micelles of 3a (C) and those at pH 2 (D).

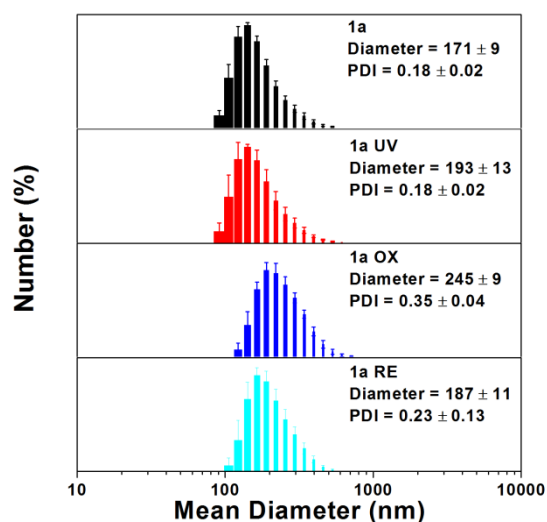


Figure S4. DLS curves of 1a nanoparticles and those upon multi-stimulation.

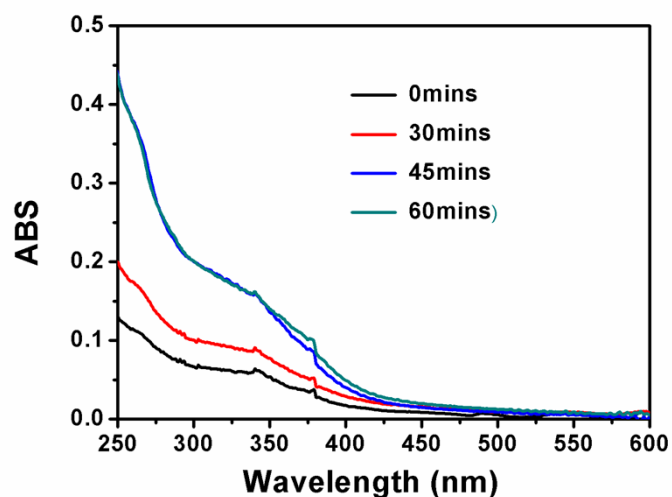


Figure S5. UV-vis spectra of aqueous solution of 0.5 mg/mL **1a** after 365 nm irradiation of different times.

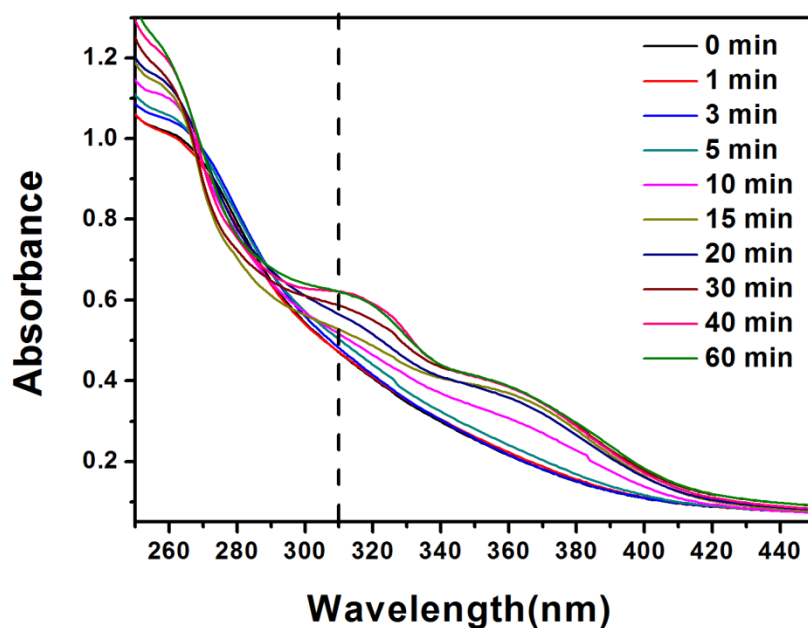


Figure S6. UV-vis spectra of aqueous solution of 1 mg/mL **4b** after 365 nm irradiation of different times. Note: The photolysis ratio of **4b** was calculated by ^1H NMR and/or from the change of the absorbance at 310 nm at different times according to our previous publications.^{1,2}

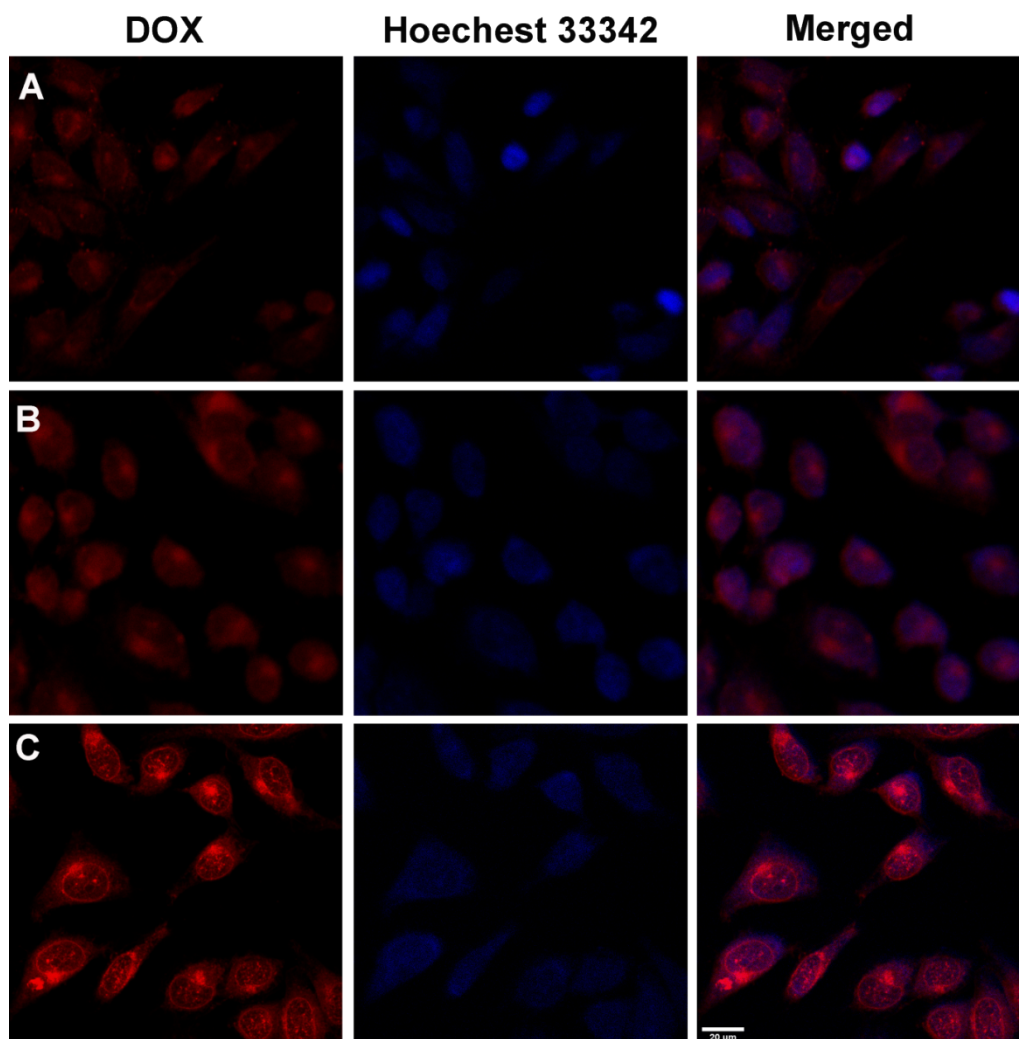
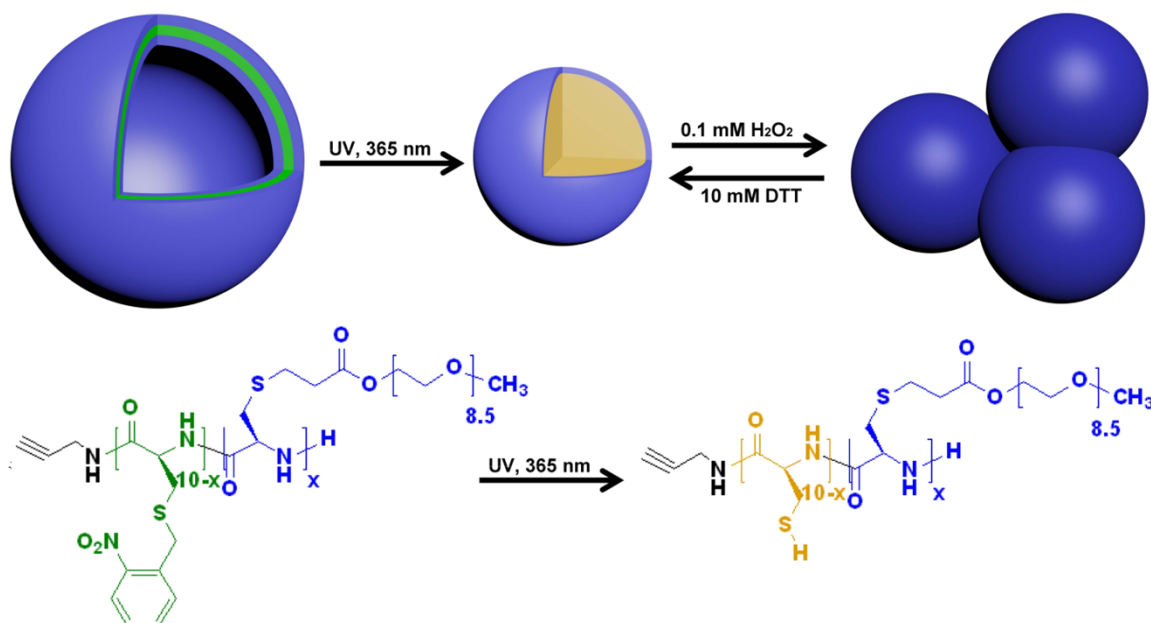


Figure S7. CLSM images of HeLa cells incubated with the DOX-loaded nanoparticles at the predetermined time intervals of 1 h (A), 2 h (B), and 4 h (C).



Scheme S1. The photo- and redox-sensitive self-assembly of 1a nanoparticles in aqueous solution.

Notes and references

1 G. Liu and C-M. Dong, *Biomacromolecules*, 2012, **12**, 1573-1583.

2 G. Liu, L. Zhou, Y. Guan, Y. Su and C-M. Dong, *Macromol. Rapid. Commun.*, 2014, **35**, 1673-1678.