## **Supporting Information**

## Controllable Fabrication of the Novel pH-, Thermo-, and Light-Responsive Supramolecular Dendronized Copolymers with Dual Self-Assembly Behavior

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Corresponding Authors:Chang\_anyang@163.com (ChangAn Yang), <u>chenxiaobo@umkc.edu</u>, zhoucongsh@126.com Figure S1-S10 and Table S1.





**Figure S1.** <sup>1</sup>H NMR spectra of G1-Azo-CHO (a),N-(4-Vinylpheny) phthalimide(b), 4-VBAHS (c) and the polymer PVBAHS (d) and in D<sub>2</sub>O.



Figure S2. FT-IR image of 4-VBAHS (a) and PVBAHS (b).



**Figure S3.** The optical images of turbidity transitions solution of monomer G1-Azo-CHO (a) and copolymer (b).



Figure S4. Concentration effcts on the LCST of monomersolution.







**Figure S5.** The chemical shift of benzylamine (a) and the chemical shift (b) of model system (the model self-assembly system were form by use the benzylamine to assembly with G1-Azo-CHO to illustrate the formation of hydrogen and imine bonds); The integral of chemical shift details of model system were in (c-f); (g) the details of chemical shift of monomer and the model system in pH=1.3,6.2,10.1.



Figure S6. FT-IR spectrum of the model system in different pH.



**Figure S7.** (a) The effect of pH on the LCST of dendronized copolymers aqueous solution before UV irradiation; (b)The effect of pH on the LCST of dendronized copolymers aqueous solution after UV irradiation.



**Figure S8.** Absorption spectrums of the self-assembly polymer at the molar ratio 0.2:1 (a), 0.4:1 (b), 0.8:1 (c), 1:1 (d).



**Figure S9.** HPLC outflow curve of the copolymer (a)  $P_{0.2}$  and (b)  $P_{0.4}$  solution at the molar ratio 1:1(CH<sub>3</sub>OH/H<sub>2</sub>O: 80/20).



**Figure S10.** Thermo-responsive of self-assembly polymers with different  $CHO/NH_2$  ratio in heating (solid line) and cooling (dot line) process.

Samples	Concentration of	Mw of the	$n_{CHO}/n_{NH2}$ -	LCST (°C)	
	total materials	copolymers		heating	cooling
	(mg/mL)	(g/mol)	(11101/11101)*		
1	5.0	7.1× 10 <sup>4</sup>	1:1	54.4	54.1
2	5.0	$5.9 \times 10^4$	0.8:1	53.5	53.5
3	5.0	$4.7 \times 10^4$	0.6:1	50.3	49.0
4	5.0	$3.5 \times 10^4$	0.4:1	46.5	46.1
5	5.0	$2.4 \times 10^4$	0.2:1	45.6	45.3

**Table S1.** Raw material molar ratios for self-assembly of supramoleculardendronized copolymers