Supplementary material

Multiresponsive Cellulose Nanocrystal Cross-Linked Copolymer Hydrogels for the Controlled Release of Dyes and Drugs

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Fig. S1. Photographs of the typical hydrogels at 52 °C. From left to right: hydrogels

of PAAc-co-PAAm, PAAc/PAAm/CNC₁, PAAc/PAAm/CNC₂, and

PAAc/PAAm/CNC₃.



Figure S2. DSC curves of CNCs and the PAAc-co-PAAm and PAAc/PAAm/CNC₂ hydrogels in the whole temperature range.



Fig. S3. Photographs of the PAAc-co-PAAm-DCNC₂ hydrogels at 52 °C and 2 °C.



Fig. S4. Photographs of (a) PAAc-co-PAAm/CNC hydrogels with varying mole ratios of AAc to AAm at a fixed CNC concentration of 2%, and (b) PAAc-co-PAAm and PAAc/PAAm/CNC₂ in the initial state and after a heating–cooling process in the final state.



Fig. S5. (a) Reversibility of the PAAc/PAAm/CNC₂ hydrogel after 10 cycles at 37 and 2 °C; (b) SEM image of PAAc/PAAm/CNC₂ hydrogels after 10 cycles of cooling and heating.



Fig. S6. Calibration curves of (a) MB and (b) MO in aqueous solutions, and the MB

(c) and MO (d) solutions before (left) and after (right) adsorption.



Fig. S7. (a) Photographs of PAAc/PAAm/CNC₂ hydrogels in water (top) and in different salt solutions (bottom). (b) Shrinkage ratios of PAAc/PAAm/CNC₂ hydrogels in different salt solutions to those in pure water. All systems were equilibrated at room temperature for 2 days.



Fig. S8. Calibration curve of DOX-HCl in aqueous solution.



Fig. S9. Cross-sectional SEM image of the dried DOX-HCl@PAAc/PAAm/CNC₂ hydrogel after 10 cycles of cooling-induced adsorption and heating-induced release by alternating the temperature between 2 °C and 37 °C.