SUPPLEMENTARY MATERIALS

Poly(*N,N’*-diethylacrylamide)-based thermoresponsive hydrogels with double network structure

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**Figure S1.** (a) Temperature dependences of swelling ratio and (b) *p*-fraction for DNA1 hydrogel realized on two specimens (1st and 2nd experiment show good reproducibility).



**Figure S2.** Temperature dependence of swelling ratio for SN hydrogels. Experimental points are fitted according to Eq. (16).



**Figure S3.** Temperature dependence of swelling ratio for DNA hydrogels. Experimental points are fitted according to Eq. (16).



**Figure S4.** Temperature dependence of swelling ratio for DNM hydrogels. Experimental points are fitted according to Eq. (16).



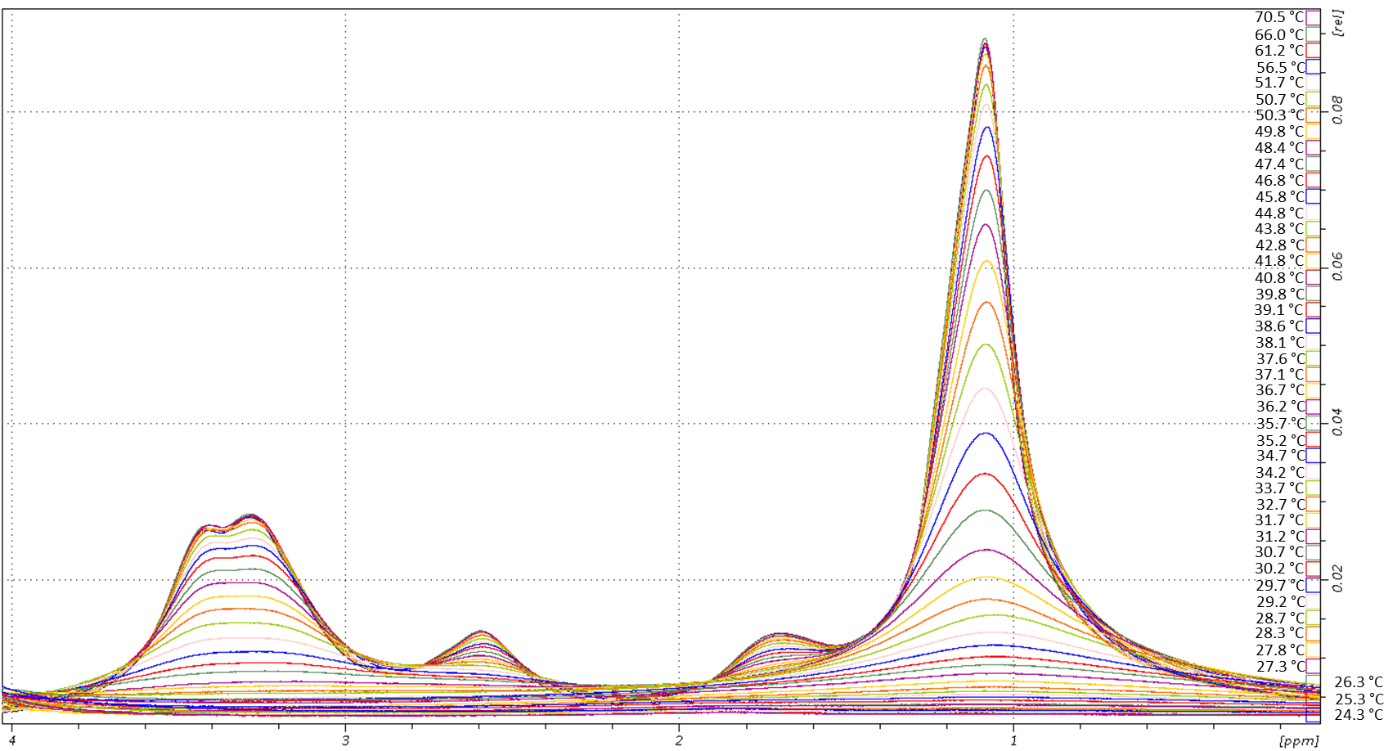
**Figure S5.** DSC curves for SN hydrogels obtained during heating.



**Figure S6.** DSC curves for DNA hydrogels obtained during heating.



**Figure S7.** DSC curves for DNM hydrogels obtained during heating.

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E

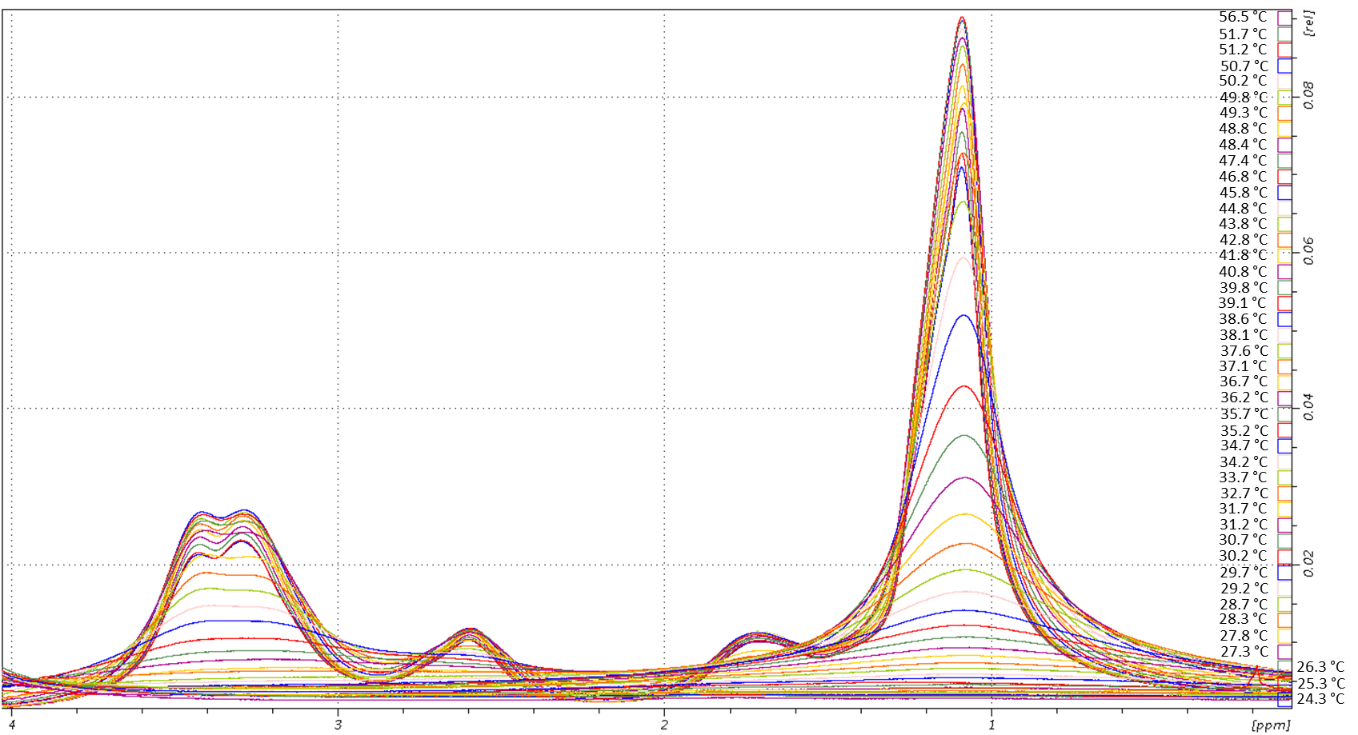
D

C

B

Increasing temperature

**Figure S8.** 1H spectra of hydrogel SN1 recorded at various temperatures. Peak assignments are explained in the text.



E

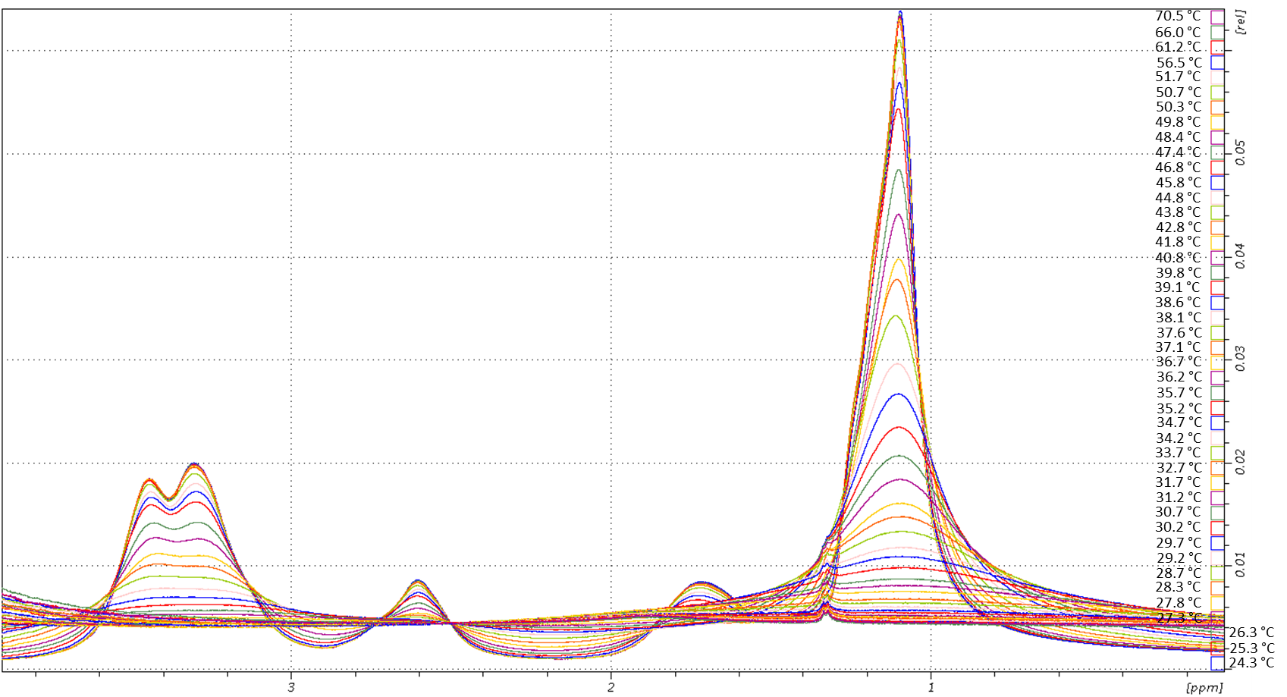
D

C

B

Increasing temperature

**Figure S9.** 1H spectra of hydrogel SN2 recorded at various temperatures. Peak assignments are explained in the text.



B

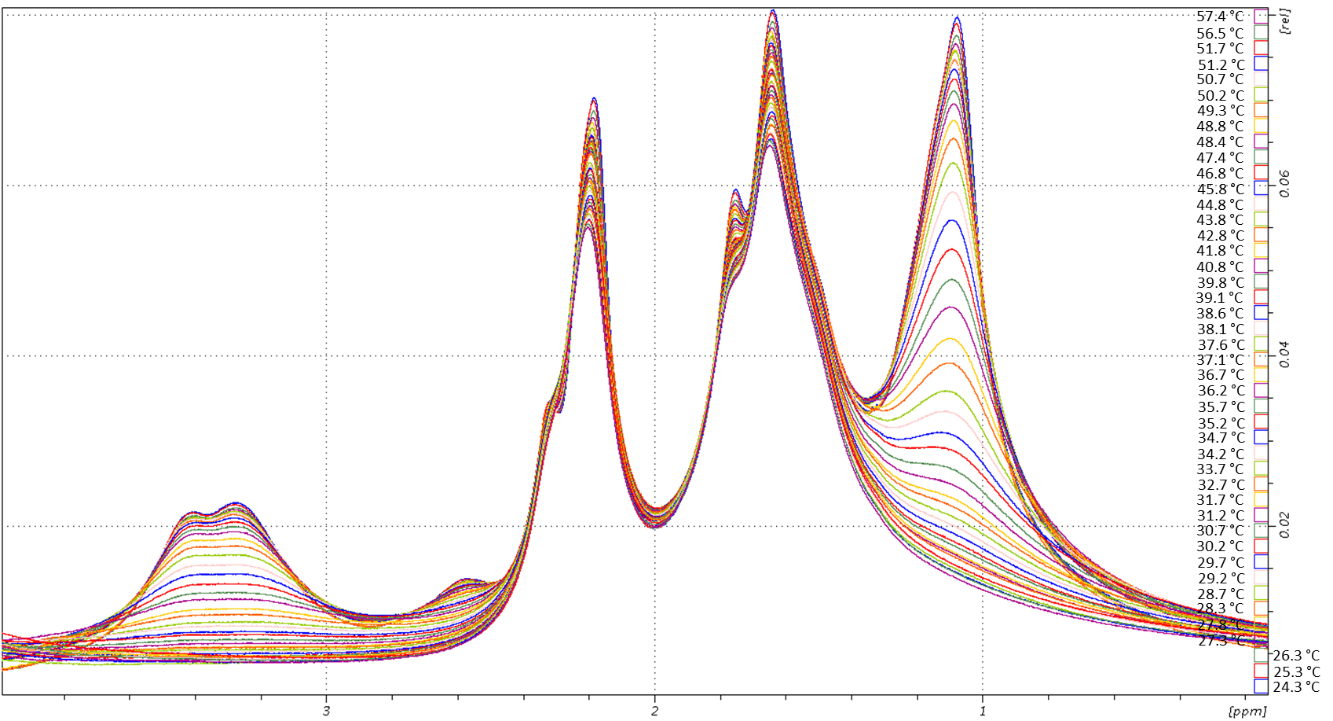
C

D

E

Increasing temperature

**Figure S10.** 1H spectra of hydrogel SN3 recorded at various temperatures. Peak assignments are explained in the text.



B

C

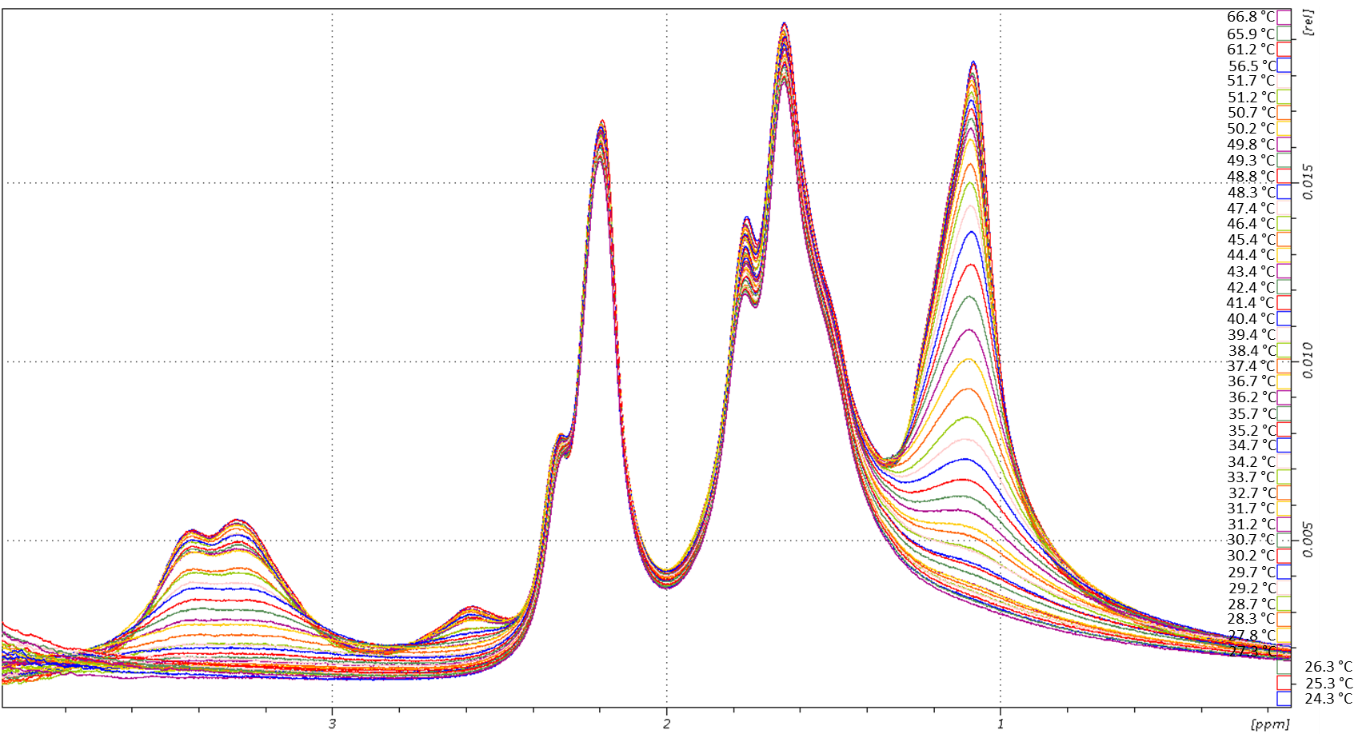
C’

D

E

Increasing temperature

**Figure S11.** 1H spectra of hydrogel DNA1 recorded at various temperatures. Peak assignments are explained in the text.



C’

D

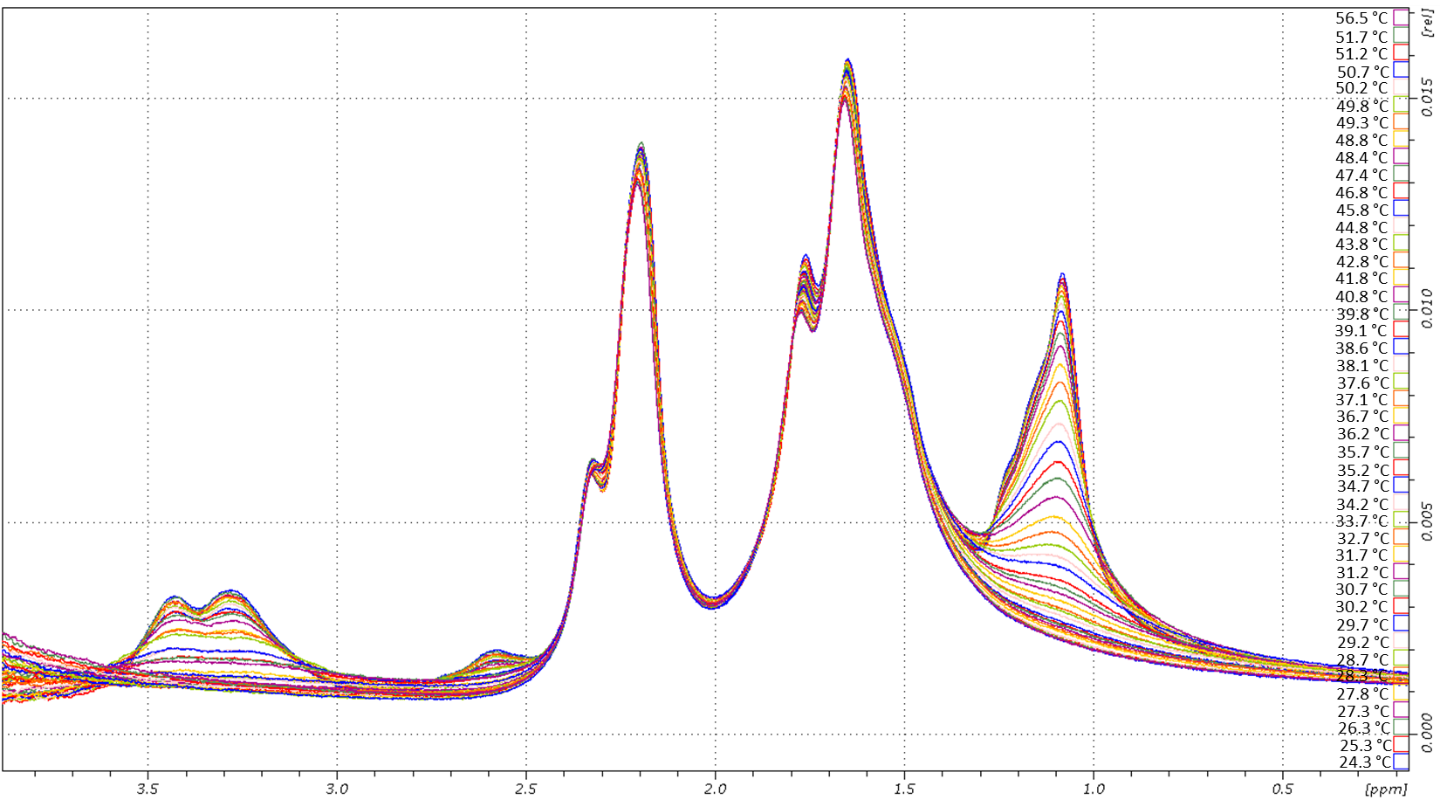
E

Increasing temperature

C

B

**Figure S12.** 1H spectra of hydrogel DNA2 recorded at various temperatures. Peak assignments are explained in the text.



Increasing temperature

E

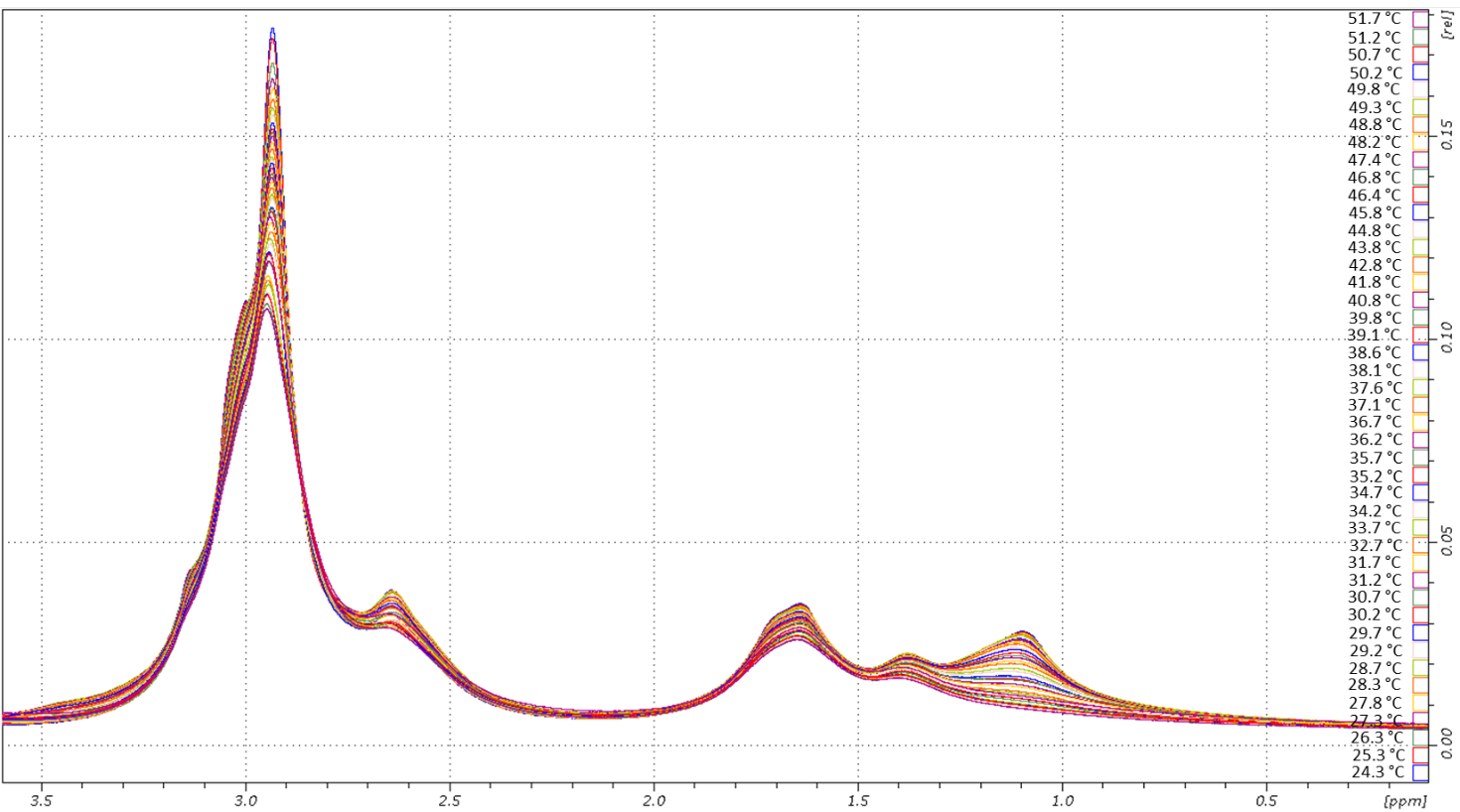
D

C’

C

B

**Figure S13.** 1H spectra of hydrogel DNA3 recorded at various temperatures. Peak assignments are explained in the text.



F

D’

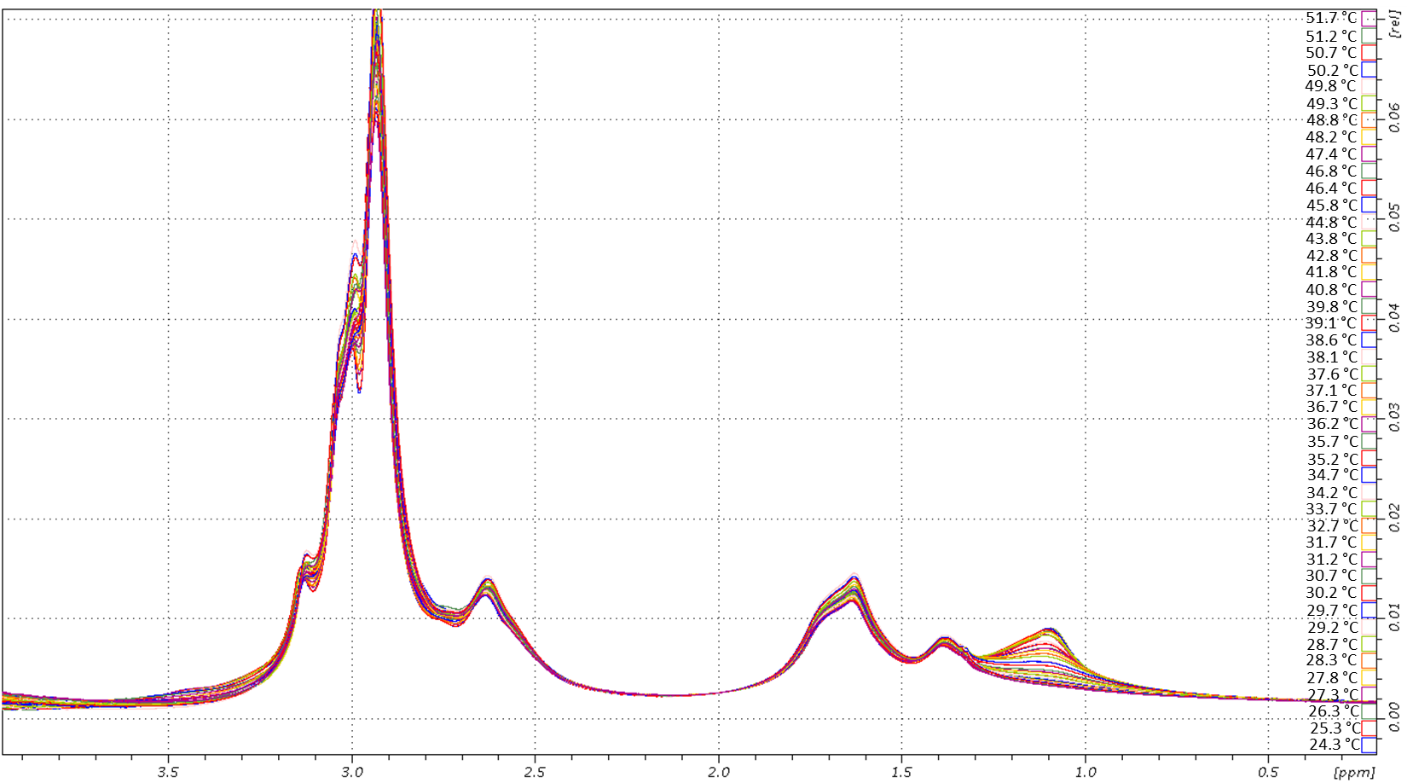
B´

C

E

Increasing temperature

**Figure S14.** 1H spectra of hydrogel DNM1 recorded at various temperatures. Peak assignment: CH3 group of PDMAAm and CH2 group of PDEAAm (B’), CH main chain group of PDEAAm (C), main chain groups CH2 of PDEAAm and CH of PDMAAm (D’), main chain group CH2 of PDMAAm (F), CH3 group of PDEAAm (E).



Increasing temperature

E

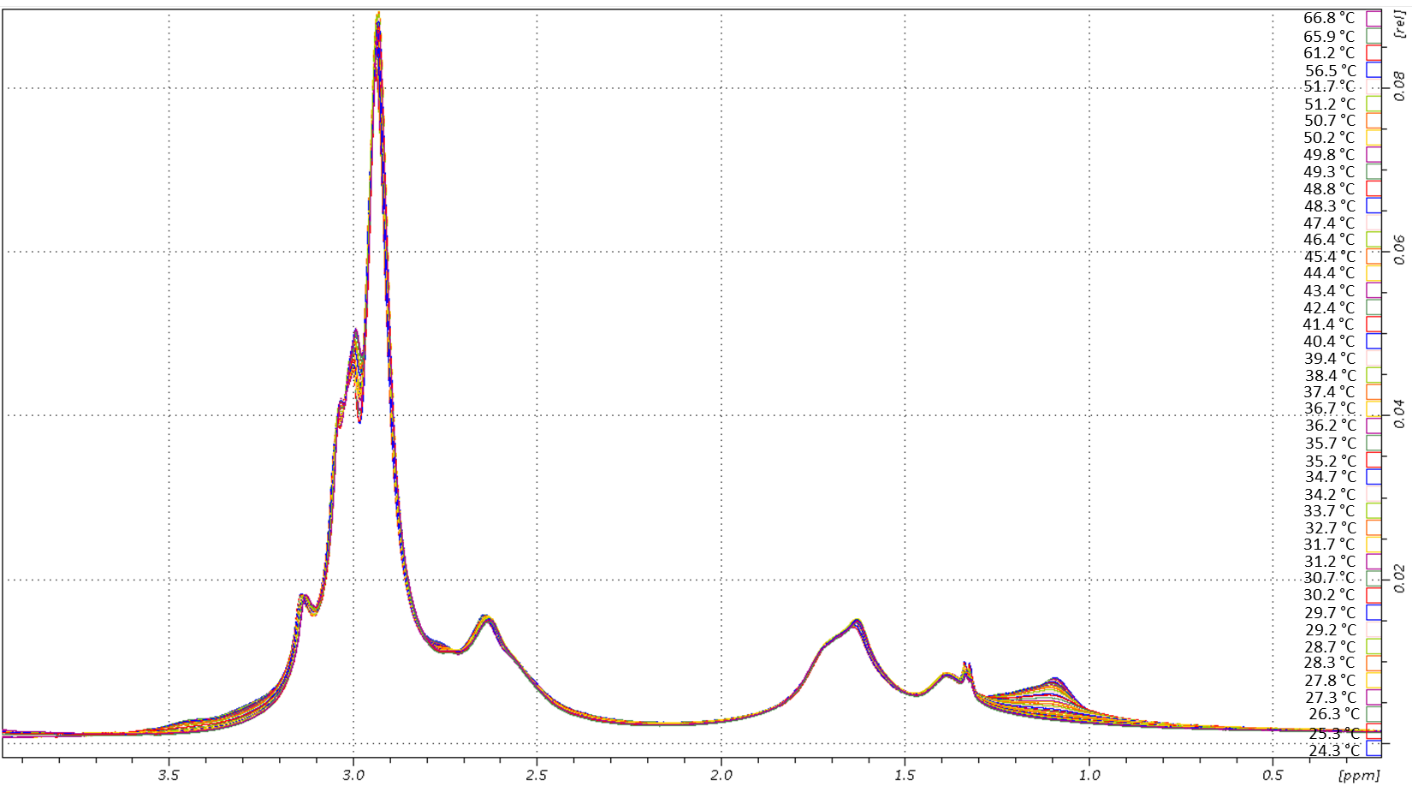
F

D’

C

B´

**Figure S15.** 1H spectra of hydrogel DNM2 recorded at various temperatures. Peak assignment: CH3 group of PDMAAm and CH2 group of PDEAAm (B’), CH main chain group of PDEAAm (C), main chain groups CH2 of PDEAAm and CH of PDMAAm (D’), main chain group CH2 of PDMAAm (F), CH3 group of PDEAAm (E).



Increasing temperature

F

D’

B´

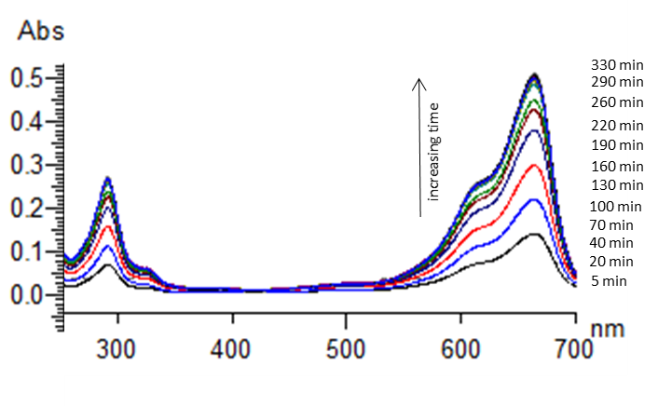
C

E

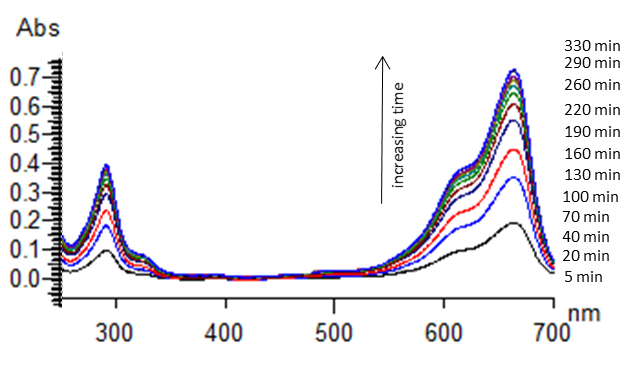
**Figure S16.** 1H spectra of hydrogel DNM3 recorded at various temperatures. Peak assignment: CH3 group of PDMAAm and CH2 group of PDEAAm (B’), CH main chain group of PDEAAm (C), main chain groups CH2 of PDEAAm and CH of PDMAAm (D’), main chain group CH2 of PDMAAm (F), CH3 group of PDEAAm (E).



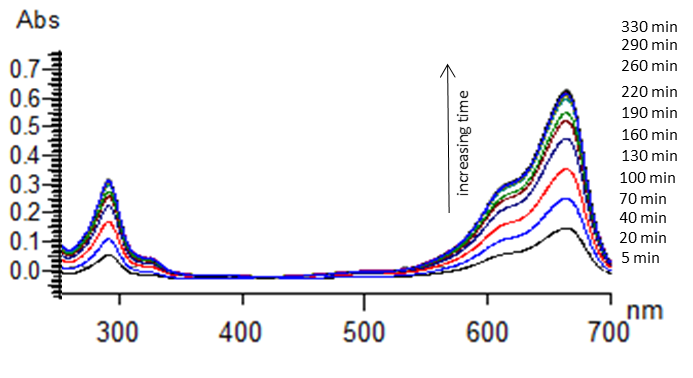
**Figure S17.** Temperature dependences of *p*-fraction for DNM hydrogels. Experimental points are fitted according to Eq. (2).



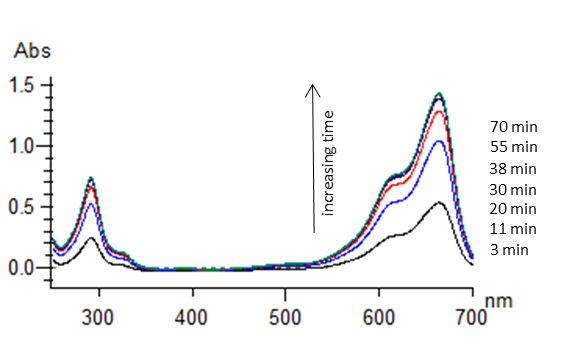
**Figure S18.** UV-Vis absorbance spectra of methylene blue released from hydrogel SN1 as a function of time.



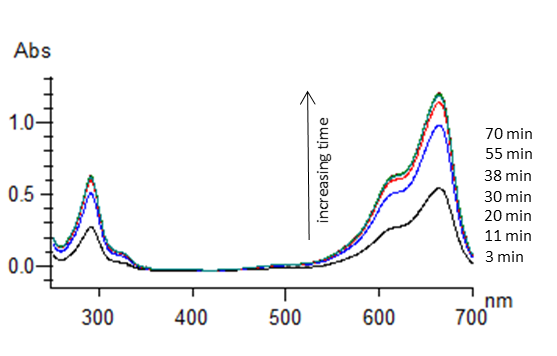
**Figure S19.** UV-Vis absorbance spectra of methylene blue released from hydrogel SN2 as a function of time.



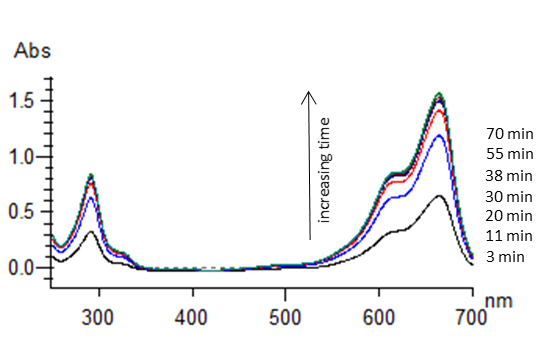
**Figure S20.** UV-Vis absorbance spectra of methylene blue released from hydrogel SN2 as a function of time.



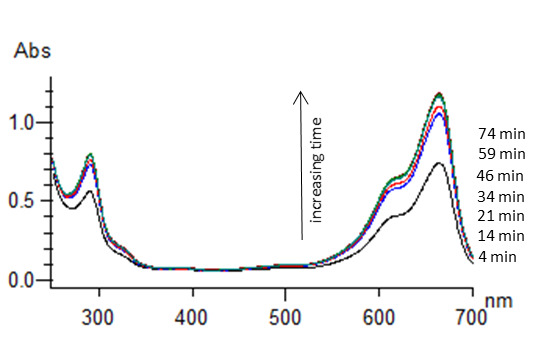
**Figure S21.** UV-Vis absorbance spectra of methylene blue released from hydrogel DNA1 as a function of time.



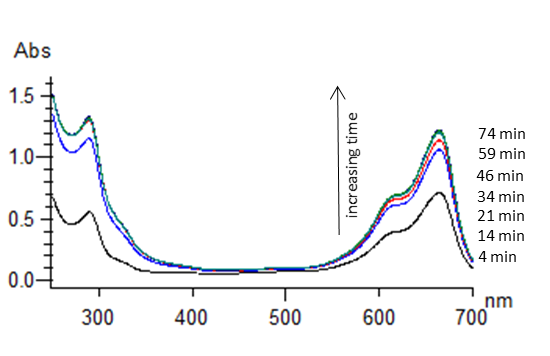
**Figure S22.** UV-Vis absorbance spectra of methylene blue released from hydrogel DNA2 as a function of time.



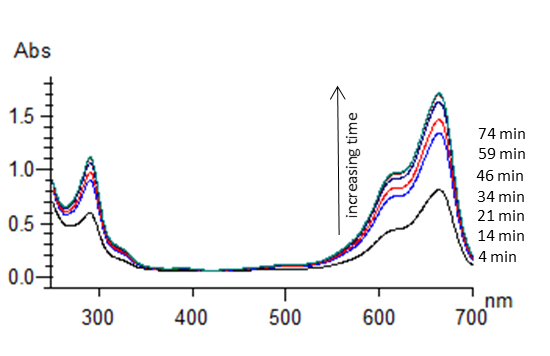
**Figure S23.** UV-Vis absorbance spectra of methylene blue released from hydrogel DNA3 as a function of time.



**Figure S24.** UV-Vis absorbance spectra of methylene blue released from hydrogel DNM1 as a function of time.



**Figure S25.** UV-Vis absorbance spectra of methylene blue released from hydrogel DNM2 as a function of time.



**Figure S26.** UV-Vis absorbance spectra of methylene blue released from hydrogel DNM3 as a function of time.



**Figure S27.** Time dependence of MB concentration for the release process for SN hydrogels. Experimental points are fitted according to Eq. (4).



**Figure S28.** Time dependence of MB concentration for the release process for DNA hydrogels. Experimental points are fitted according to Eq. (4).



**Figure S29.** Time dependence of MB concentration for the release process for DNM hydrogels. Experimental points are fitted according to Eq. (4).