Electronic Supplementary Material (ESI) for Soft Matter.

Supplementary Information

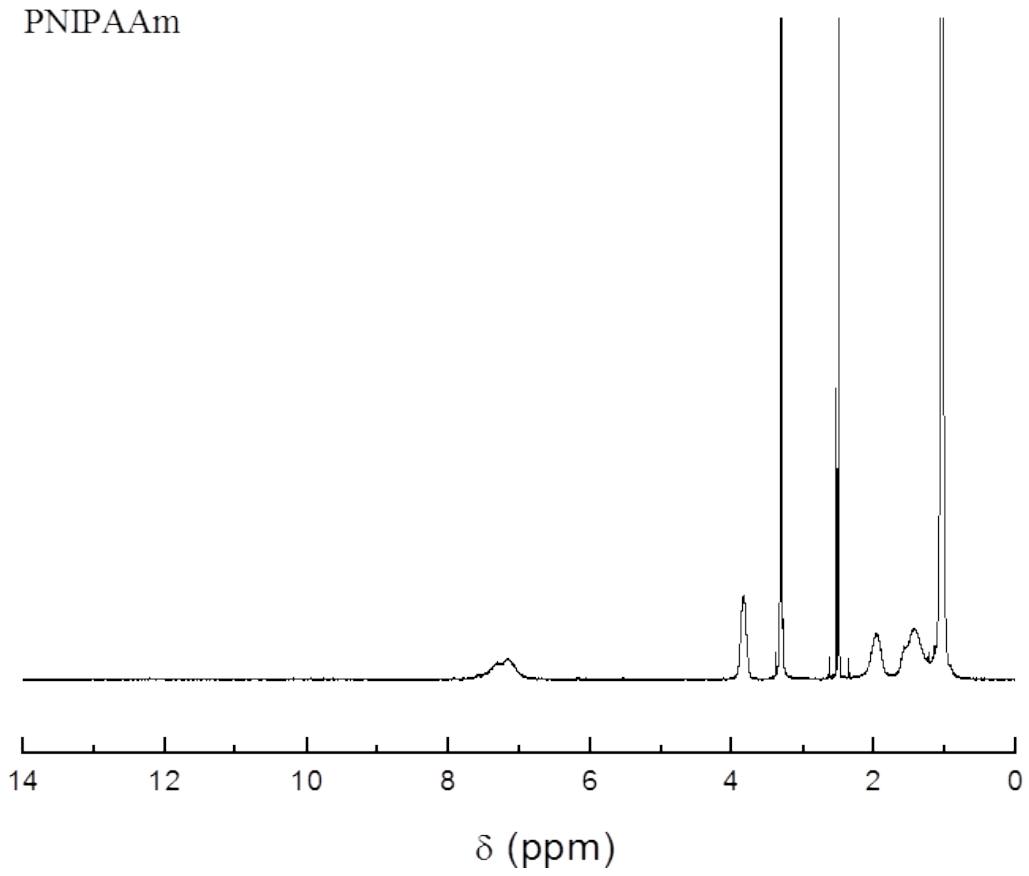


Figure S1 ^{1}HNMR spectrum of PNIPAAm.

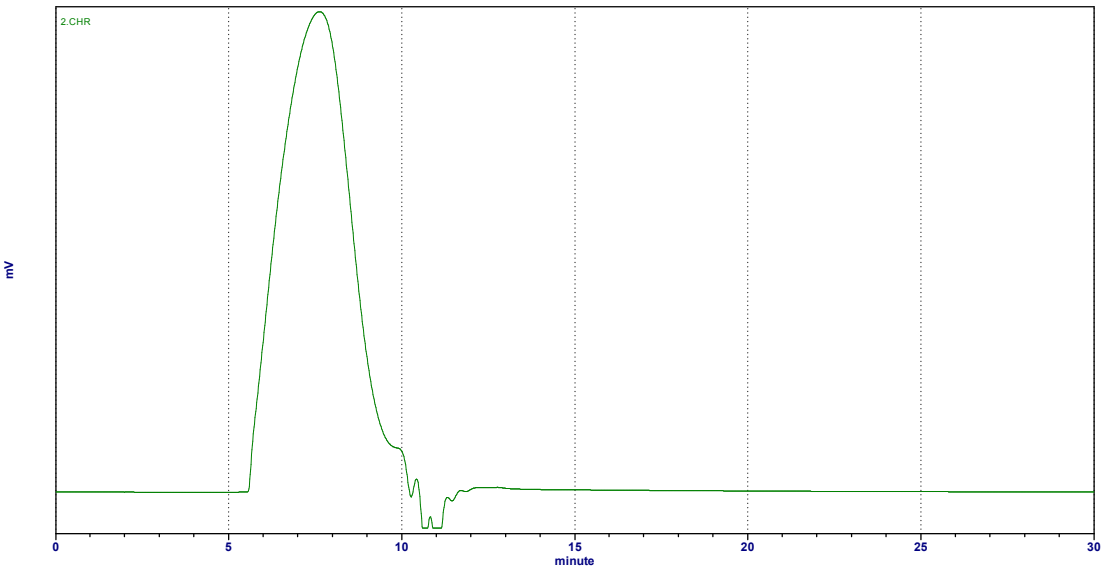


Figure S2 GPC curve of PNIPAAm.

	min	M.W.	Mw	PDI
1	7.410	27473	34625	1.2603

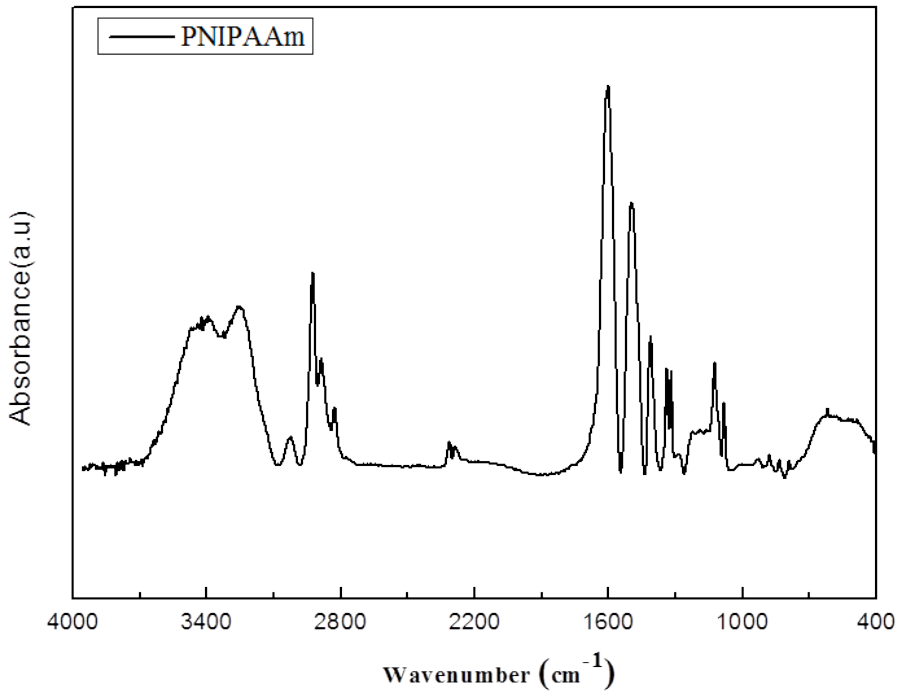


Figure S3 FTIR spectrum of PNIPAAm.

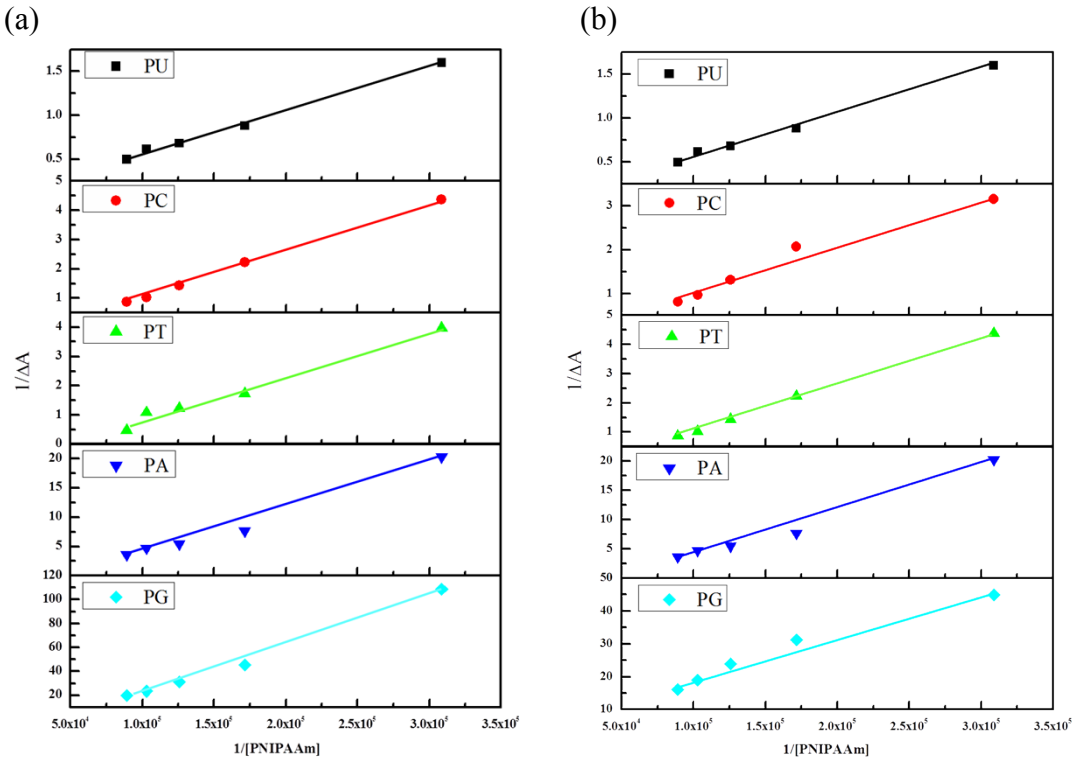


Figure S4 Reciprocal of absorbance area change at 260 nm plotted with respect to the reciprocal of the concentration of PNIPAAm at (a) 25 and (b) 45 °C with linear fitting according to the Eq. (3).

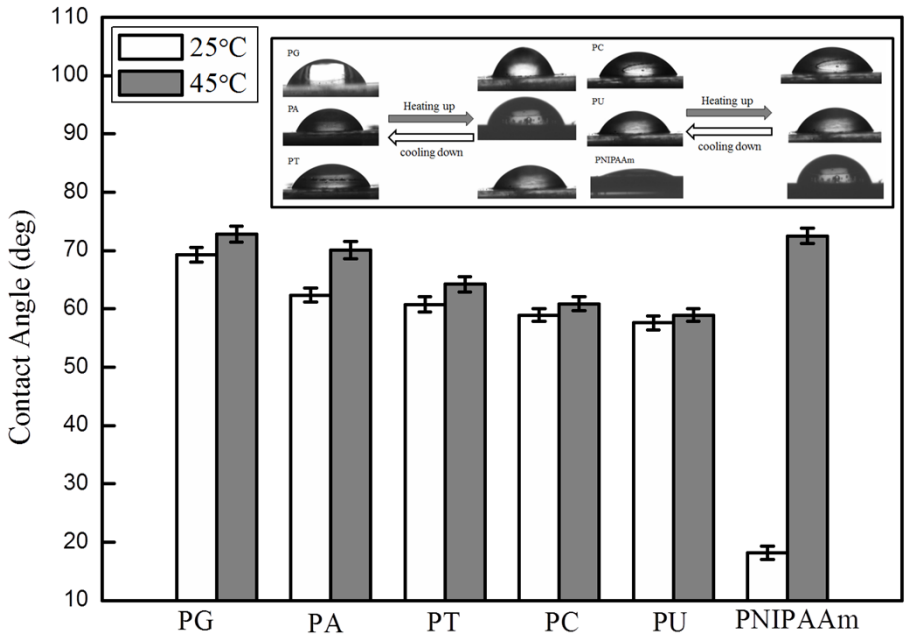
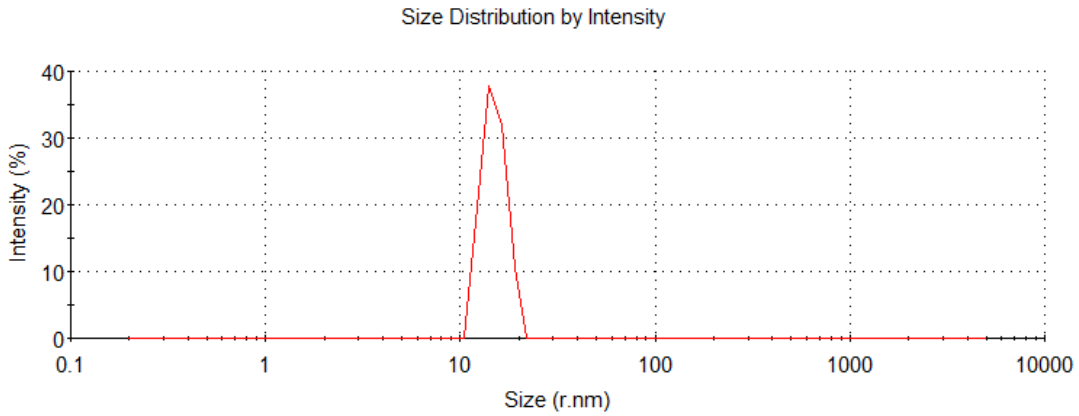


Figure S5 SWCAs of PNIPAAm, PG, PA, PT, PC, and PU measured at 25 and 45 °C, as well as photographic images of water droplets on their surfaces.

PNIPAAm is soluble in water at 25 °C, because its amido groups readily form HBs with surrounding water molecules, resulting in a low SWCA (18 ± 3 °C). Intramolecular HBs between the amido groups and increasingly strong hydrophobic interactions among isopropyl groups prevail at 45 °C, resulting in the SWCA increasing to 73 ± 3 °C. The five kinds different nucleobases in the PNSCs significantly suppressed the thermoresponsive behavior of the PNIPAAm, because of BMHBs between the amido group and the nucleobases. (Supplementary Information; Figure S5) The decrease in hydrophilicity of the PNSCs can be explained by considering that strong BMHBs were formed between the hydrophilic groups of the PNIPAAm and the nucleobases, resulting in the hydrophobic groups being presented predominantly on the surface. Moreover, our results revealed that the SWCA of the PG surface was higher than those of the other PNSC films at 25 °C, suggesting a higher degree of hydrophobicity in that system because it featured the strongest BMHBs among our tested PNSCs. The three BMHBs between the PNIPAAm and each G unit weakened slightly at 45 °C, leading to increased hydrophilicity. The presence of only two BMHBs between the PNIPAAm and each of the other nucleobases significantly suppressed the thermoresponsive hydrophobicity of the polymer. The thermoresponsive hydrophobicity of the PNIPAAm disappeared completely in the presence of PC and PU, even through the BMHBs in PC and PU were weaker than those of the other PNSCs. These results imply that the BMHBs could change the surface morphology of the polymer and influence its SWCAs.

(a)



(b)

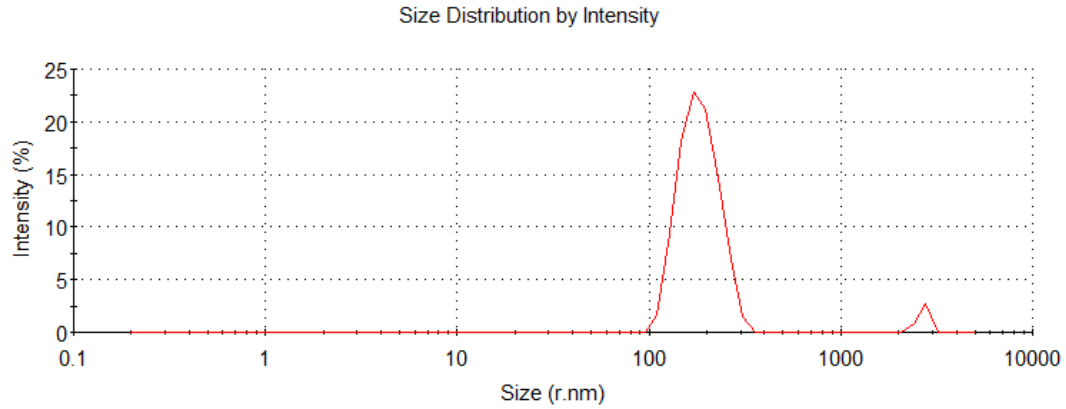


Figure S6 Particle size distribution of PNIPAAm, measured by DLS, at (a)25 and (b)45 °C.

Table S1 *I–V* Characteristics of films of PNSCs under various electric fields.

Sample	Temperature	Current (nA) at selected voltages					
		–6 V	–4 V	–2 V	2 V	4 V	6 V
PG	30 °C	–443.1	–322.4	–159.9	139.1	301.5	441.7
	40 °C	–420.7	–308.2	–151.1	128.9	264.1	412.1
	50 °C	–401.9	–273.7	–148.0	123.5	253.8	385.2
	60 °C	–382.9	–266.9	–142.8	113.8	246.9	362.1
	70 °C	–361.7	–248.7	–128.6	104.4	228.7	344.8
PA	30 °C	–312.5	–217.0	–112.3	92.1	193.9	300.3
	40 °C	–288.7	–196.1	–105.3	86.3	186.7	276.3
	50 °C	–243.2	–165.8	–85.7	69.6	150.9	231.7
	60 °C	–210.8	–146.9	–80.4	60.5	125.7	192.4
	70 °C	–185.8	–126.6	–68.9	49.8	114.3	176.2
PT	30 °C	–125.2	–86.6	–45.1	37.8	79.4	118.7
	40 °C	–80.4	–52.6	–27.5	22.3	51.6	78.7
	50 °C	–62.7	–48.1	–21.6	18.4	38.3	58.9
	60 °C	–40.4	–27.5	–15.2	11.2	21.6	35.7
	70 °C	–20.2	–13.8	–6.8	6.1	10.6	18.9
PC	30 °C	–20.1	–13.5	–6.7	6.1	12.7	19.2
	40 °C	–10.3	–6.9	–3.5	2.8	6.1	9.4
	50 °C	–0.2	–0.13	–0.07	0.06	0.11	0.2
	60 °C	–0.2	–0.13	–0.07	0.06	0.11	0.2
	70 °C	–0.2	–0.13	–0.07	0.06	0.11	0.2
PU	30 °C	–0.2	–0.13	–0.07	0.06	0.11	0.2
	40 °C	–0.2	–0.13	–0.07	0.06	0.11	0.2
	50 °C	–0.2	–0.13	–0.07	0.06	0.11	0.2
	60 °C	–0.2	–0.13	–0.07	0.06	0.11	0.2
	70 °C	–0.2	–0.13	–0.07	0.06	0.11	0.2