Supporting Information

Enhanced Adsorption of Methylene Blue Triggered by the Phase Transition of Thermo-responsive Polymer in Hybrid Interpenetrating Polymer Network Hydrogels

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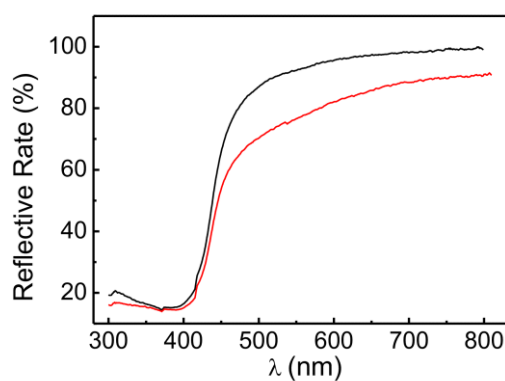
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Figure S1. The UV-Vis diffuse reflectance spectra of the hybrid alginate-Ca^{2+}/P(MEO$_2$MA-*co*-OEGMA$_{300}$)/g-C$_3$N$_4$ IPN hydrogels (red) and g-C$_3$N$_4$ (black) after freeze drying.
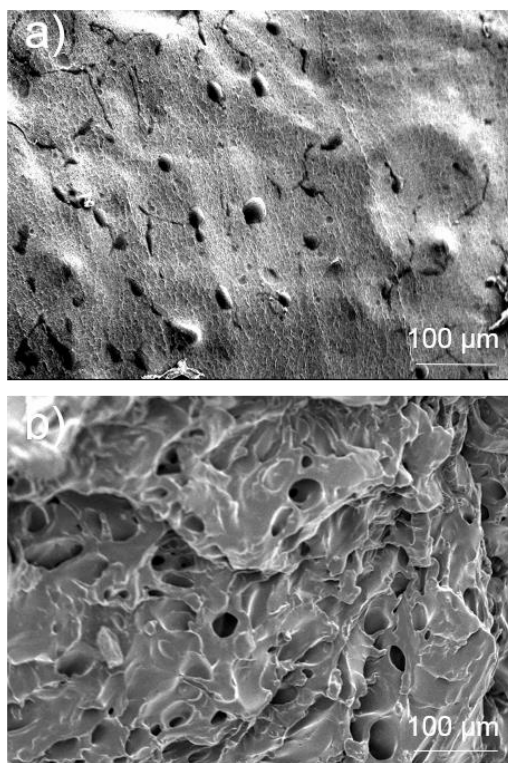
Figure S2. Cross-section SEM images of (a) the alginate-Ca^{2+}/P(MEO$_2$MA-*co*-OEGMA$_{300}$) IPN hydrogels and (b) hybrid alginate-Ca^{2+}/P(MEO$_2$MA-*co*-OEGMA$_{300}$)/g-C$_3$N$_4$ IPN hydrogels at 45 °C (TT).

Adsorption kinetics model for ionic dyes into hybrid IPN hydrogels

In order to better understand the adsorption behavior of the hybrid hydrogels, the adsorption kinetics are fitted by two kinetic models: the pseudo-first-order model by Lagergren (Eq. S1), and pseudo-second-order model (Eq. S2). These two models are used to evaluate the experimental adsorption data of methylene blue into hybrid hydrogels.

$$\lg(q_e - q_t) = \lg q_e - \frac{k_1}{2.303}t \qquad (S1)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \qquad (S2)$$

In which q_e and q_t represent the amounts of the dye adsorbed at equilibrium (mg g^{-1}) at time t, respectively. k_1 is the rate constant of pseudo-first-order kinetic (min^{-1}), and k_2 is the rate constant of pseudo-second-order kinetic (g (mg min)$^{-1}$). The

experimental data presented in Figure S1 are fitted linearly by using Eq. S1 and Eq. S2.
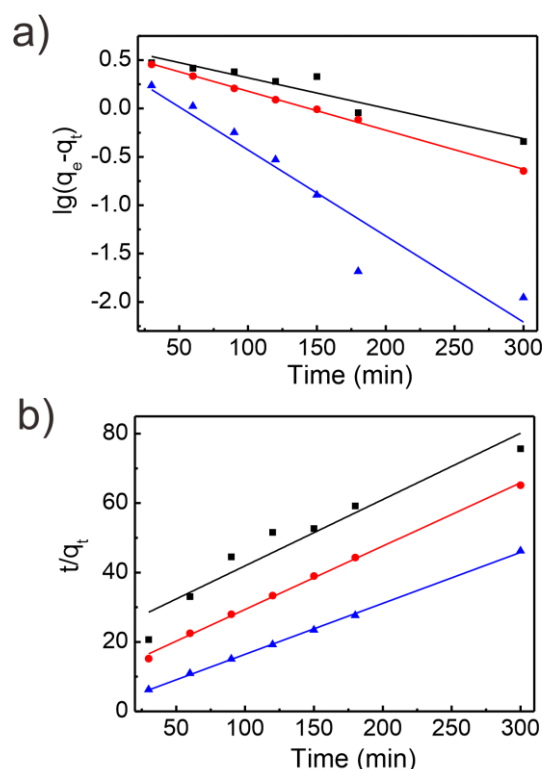

Figure S3. Comparison of adsorption kinetic models of methylene blue in the alginate-Ca^{2+}/ P(MEO$_2$MA-*co*-OEGMA$_{300}$) IPN hydrogels at 35 °C (black), the hybrid alginate-Ca^{2+}/ P(MEO$_2$MA-*co*-OEGMA$_{300}$)/g-C$_3$N$_4$ IPN hydrogels at 35 °C (red) and 45 °C (blue). (a) Linear dependence of $\lg(q_e - q_t)$ on t based on the pseudo-first-order, (b) Linear dependence of t/q_t on t based on the pseudo-second-order model.

The constants corresponding to the kinetic models are included in Table S1. As Table S1 shows, the theoretical q_e values estimated from the pseudo-second-order model were very close to the experimental values for all composite hydrogels, the correlation efficient R^2 being also high. These results show that the kinetic behavior of the hydrogel-adsorbed methylene blue dye follows a quasi-second-order kinetic equation. In addition, the effect of the temperature on the adsorption performance of hybrid alginate-Ca^{2+}/P(MEO$_2$MA-*co*-OEGMA$_{300}$)/g-C$_3$N$_4$ hydrogels was studied. As depicted in Figure 7, the adsorption rate of the hybrid hydrogel adsorbent was slightly enhanced

with increasing temperature from 30 to 45 °C. On the basis of the above results, the hybrid hydrogel can play a role as an excellent adsorbent not only for its efficient adsorption behavior on cationic aromatic dyes via strong π-π stacking interaction and ionic interaction, but also for the collapse of P(MEO$_2$MA-*co*-OEGMA$_{300}$) causes the more porous structure in the hybrid IPN network when the temperature is increased to its transition temperature (TT, 45 °C).

Table S1. Kinetic parameters for methylene blue adsorption of (a) alginate-Ca^{2+}/P(MEO$_2$MA-*co*-OEGMA$_{300}$) IPN hydrogels at 35 °C; (b) the hybrid alginate-Ca^{2+}/P(MEO$_2$MA-*co*-OEGMA$_{300}$)/g-C$_3$N$_4$ IPN hydrogels at 35 °C; (c) the hybrid alginate-Ca^{2+}/P(MEO$_2$MA-*co*-OEGMA$_{300}$)/g-C$_3$N$_4$ IPN hydrogels at 45 °C.

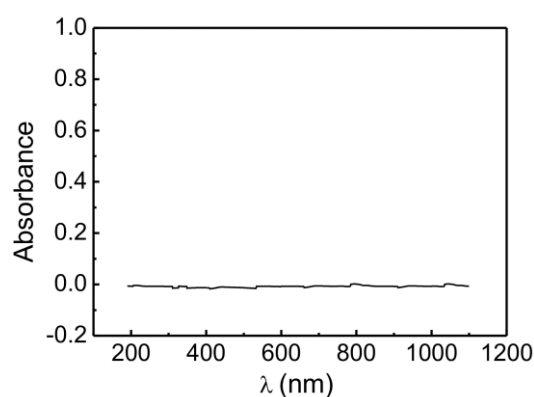| Models	Parameters	a	b	c
pseudo-first-order	R^2	0.890	0.998	0.890
	K_1	-0.003	-0.004	-0.009
	q_e	4.477	4.830	6.530
pseudo-second-order	R^2	0.909	0.998	0.991
	K_1	0.191	0.182	0.147
	q_e	4.491	5.054	6.708

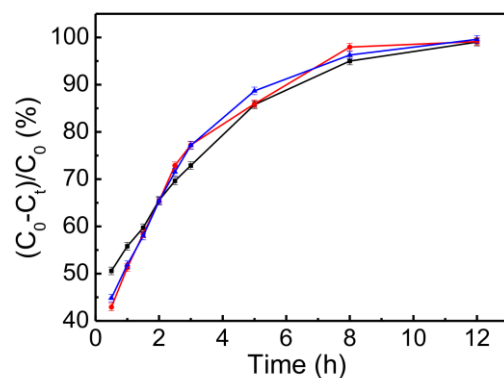**Figure S4.** UV-Vis absorption spectrum of the residual solution after the photo-degradation (12 h).

Figure S5. Three cycles of adsorption and photo-degradation measurements of the hybrid IPN hydrogels (black: 1st, red: 2nd and blue: 3rd).
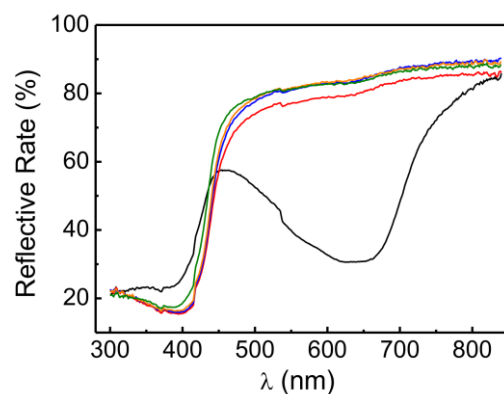

Figure S6. The UV-Vis diffuse reflectance spectra of the hybrid IPN hydrogels (red), the hybrid IPN hydrogels containing methylene blue (black) and the hybrid IPN hydrogels after the adsorption and photo-degradation measurement. The cycle number is 1 (blue), 3 (orange) and 5 (olive), respectively.
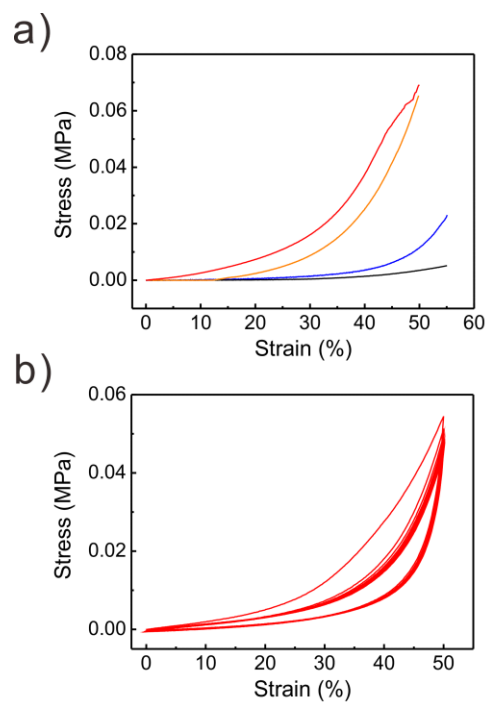
Figure S7. Mechanical properties of the hydrogels at 45 ºC (above TT): (a) Stress-strain compression curves of the P(MEO$_2$MA-*co*-OEGMA$_{300}$) hydrogels (black), alginate-Ca^{2+}/P(MEO$_2$MA-*co*-OEGMA$_{300}$) IPN hydrogels (blue) and hybrid alginate-Ca^{2+}/P(MEO$_2$MA-*co*-OEGMA$_{300}$)/g-C$_3$N$_4$ IPN hydrogels as-prepared (red) as well as after adsorption and photo-degradation (orange). (b) 10 cycles through compression with 50% strain in hybrid alginate-Ca^{2+}/P(MEO$_2$MA-*co*-OEGMA$_{300}$)/g-C$_3$N$_4$ IPN hydrogels.