

**Supporting Information**

**Agar-Reduced Graphene Oxide Selectively Adsorbs Organic Dyes and  
Strengthens Double-Network Hydrogels**

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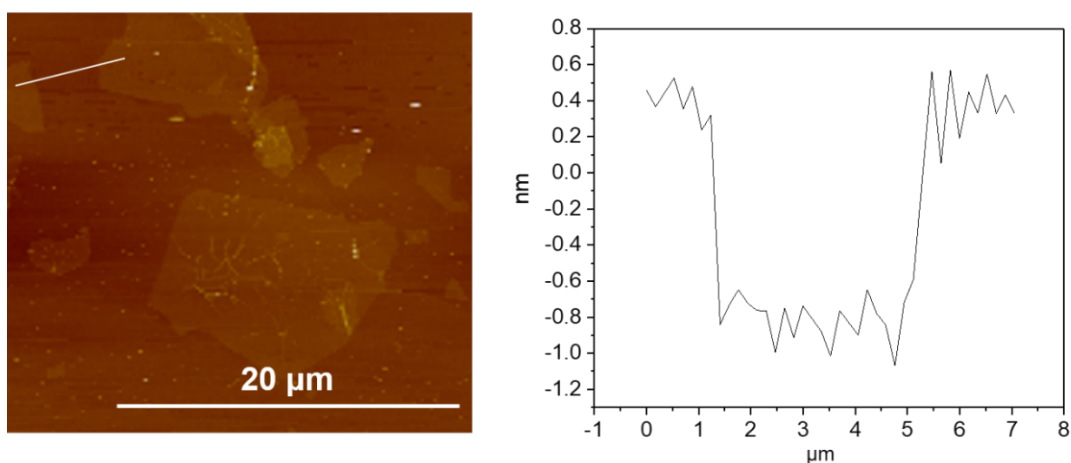
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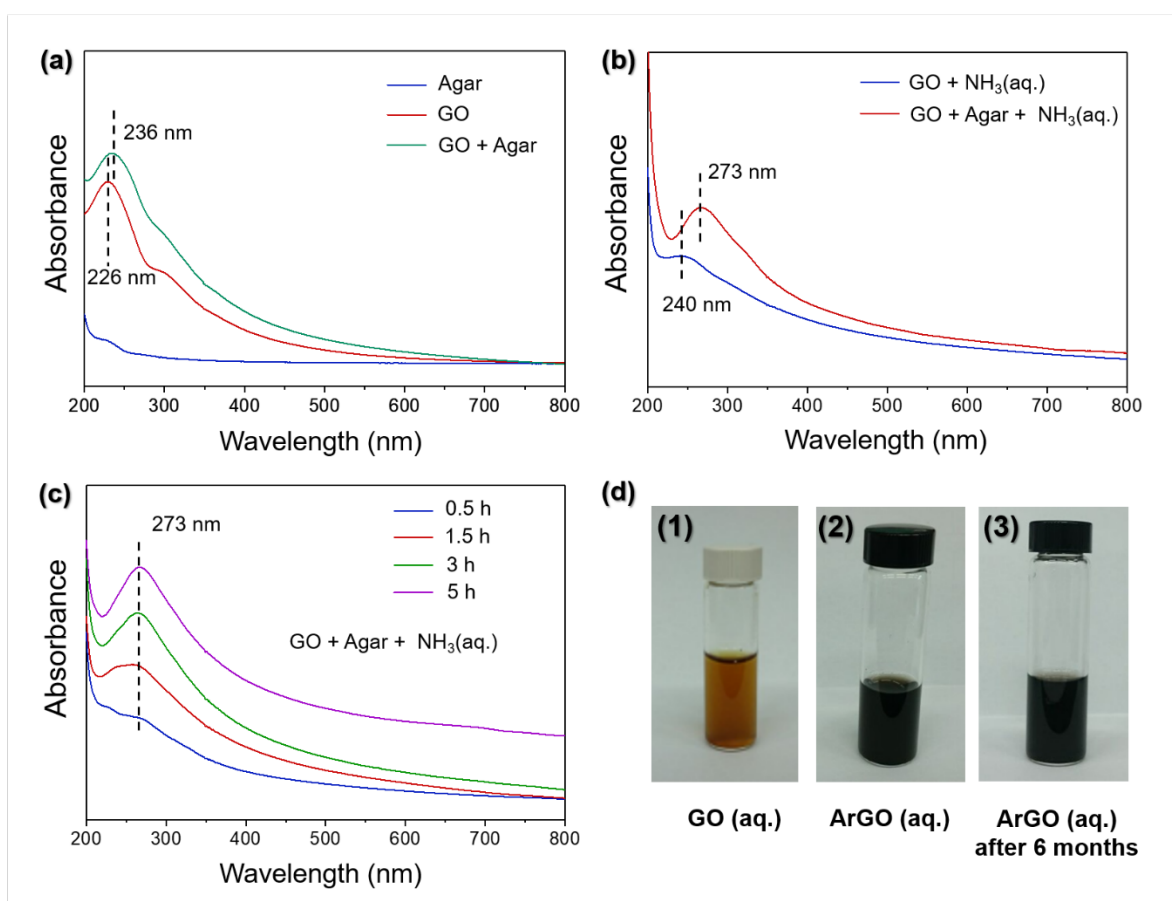
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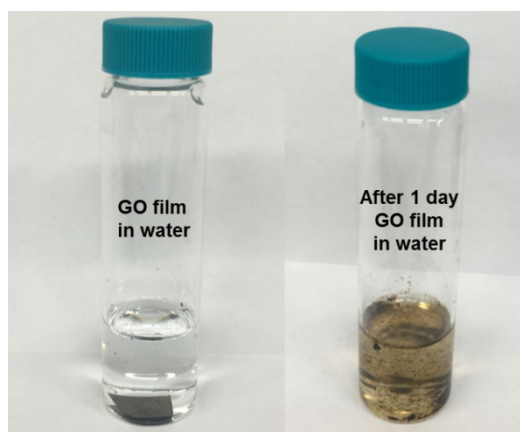


**Figure S1.** AFM image and profile data recorded for graphene oxide.

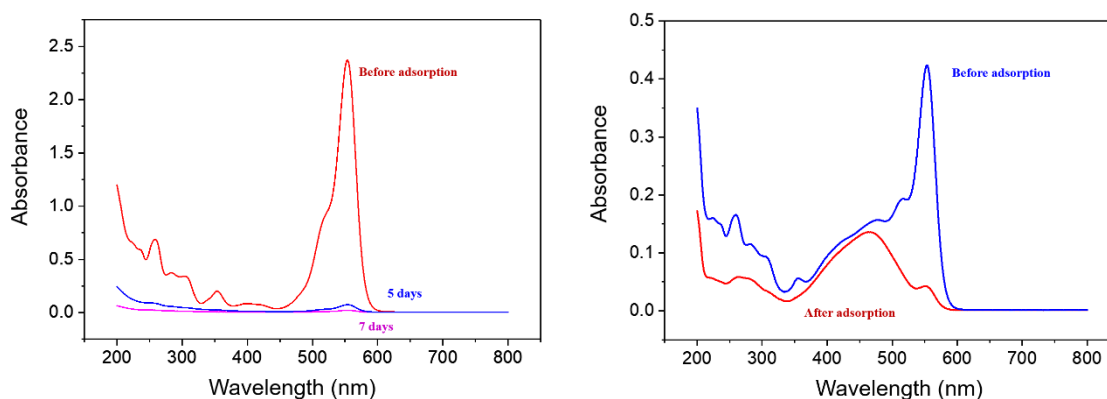


**Figure S2.** (a) UV-vis absorption spectra that were recorded for a  $2.0 \text{ mg mL}^{-1}$  aqueous dispersion of agar after being heated to  $95 \text{ }^\circ\text{C}$  for 12 h (blue); a  $0.2 \text{ mg mL}^{-1}$  aqueous dispersion of GO (red); a  $0.2 \text{ mg mL}^{-1}$  aqueous dispersion of GO with agar (agar/GO = 10 : 1 w/w) after being heated to  $95 \text{ }^\circ\text{C}$  for 12 h (green). (b) UV-vis absorption spectra that were recorded for a  $0.2 \text{ mg mL}^{-1}$  aqueous dispersion of GO and  $\text{NH}_3$  (aq.) (pH = 8 – 9) after being heated to  $95 \text{ }^\circ\text{C}$  for 3 h (blue); a  $0.2 \text{ mg mL}^{-1}$  aqueous dispersion of GO, agar (agar/GO = 10 :

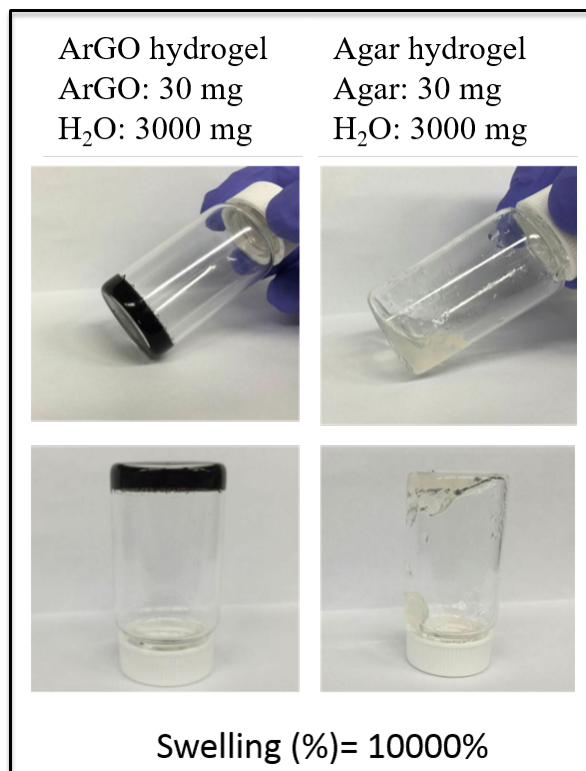
1 w/w) and  $\text{NH}_3$  (aq.) (pH = 8 – 9) after being heated to 95 °C for 3 h (red). (c) UV–vis absorption spectra that were recorded over time (indicated) for a 0.2 mg mL<sup>-1</sup> aqueous dispersion of GO, agar (agar/GO = 10 : 1 w/w) and  $\text{NH}_3$  (aq.) (pH = 8 – 9) that was kept at 95 °C. (d) Photographs of various suspensions and dispersions (indicated).



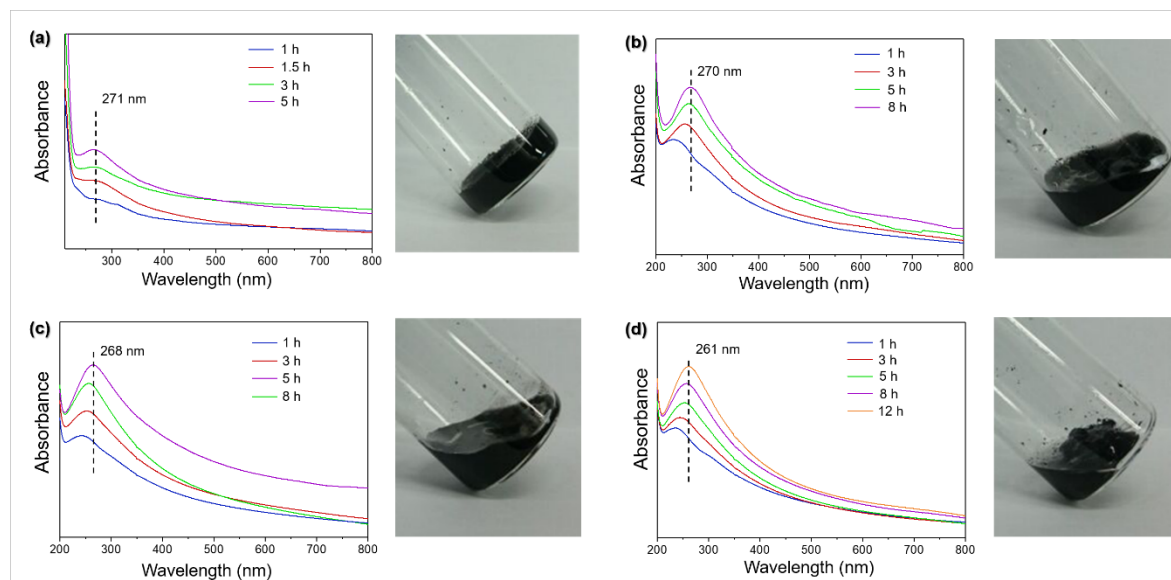
**Figure S3.** Photographs taken (left) after a film of GO was placed in water and (right) after the mixture was left standing for 1 day.



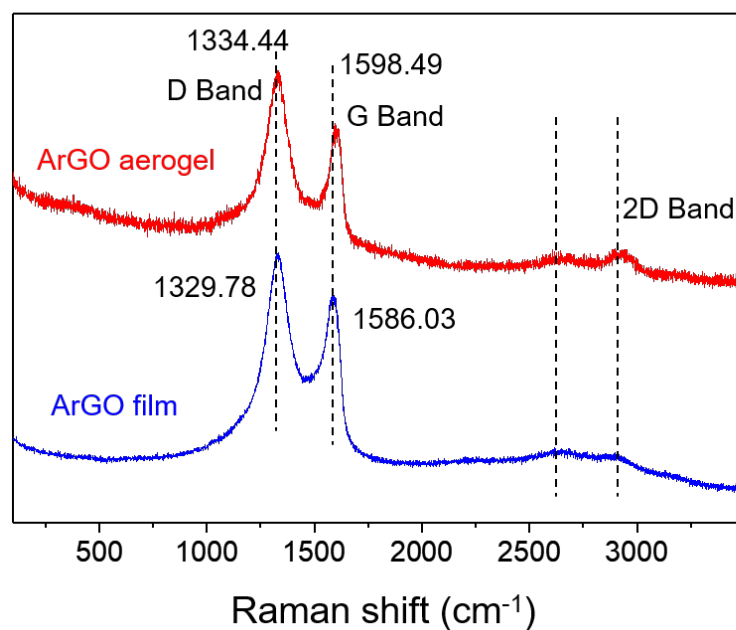
**Figure S4.** UV-vis spectra. (left) The selective adsorption of RhoB versus MO was demonstrated by soaking ArGO paper (1 mg) in an aqueous mixture of both dyes for 12 h ( $[\text{RhoB}]_0 = [\text{MO}]_0 = 1 \text{ mg L}^{-1}$ , 2 mL total volume). (right) The adsorption capacity was assessed by soaking ArGO paper (1 mg) in an aqueous solution of RhoB for 7 days ( $[\text{RhoB}]_0 = 20 \text{ mg L}^{-1}$ , 2 mL total volume).



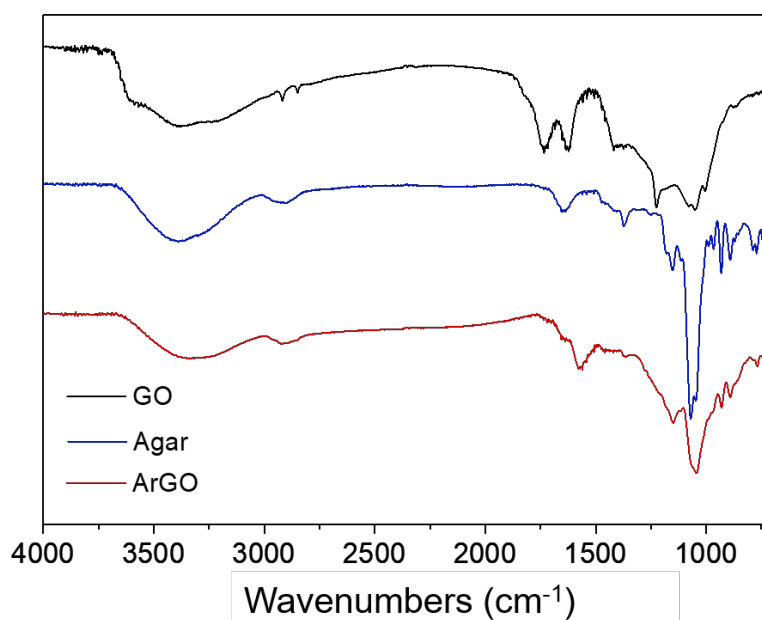
**Figure S5.** Swelling properties of an ArGO hydrogel and an agar hydrogel (indicated).



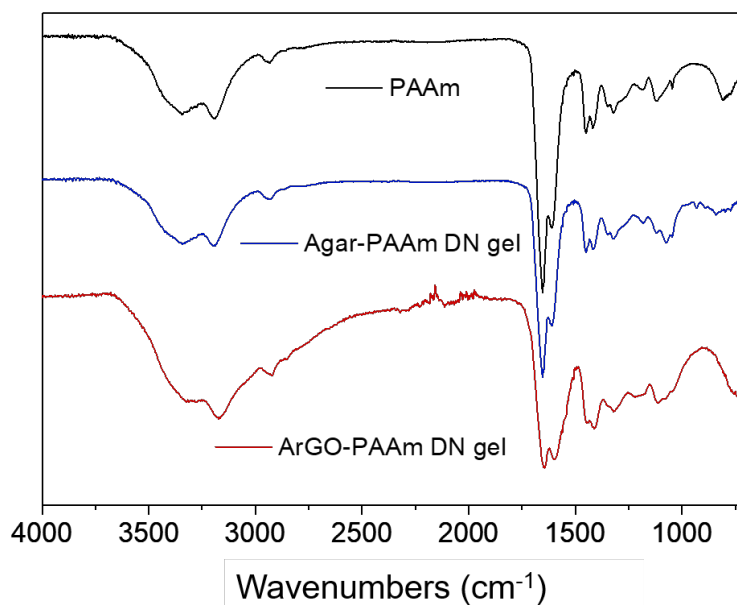
**Figure S6.** UV-vis absorption spectra that were recorded over time and photographs that were recorded over the sol-gel transition. Conditions: 1.0 mg mL<sup>-1</sup> aqueous dispersion of GO + NH<sub>3</sub> (aq.) (pH=8-9, mixed with (a) agar (agar/GO = 10 : 1 w/w), (b) agar (agar/GO = 5 : 1 w/w), (c) agar (agar/GO = 2 : 1 w/w) and (d) agar (agar/GO = 1 : 1 w/w). Each mixture was heated to 95 °C for the period indicated in the figure legends. Inset: photographs of the ArGO hydrogels that resulted from the reactions.



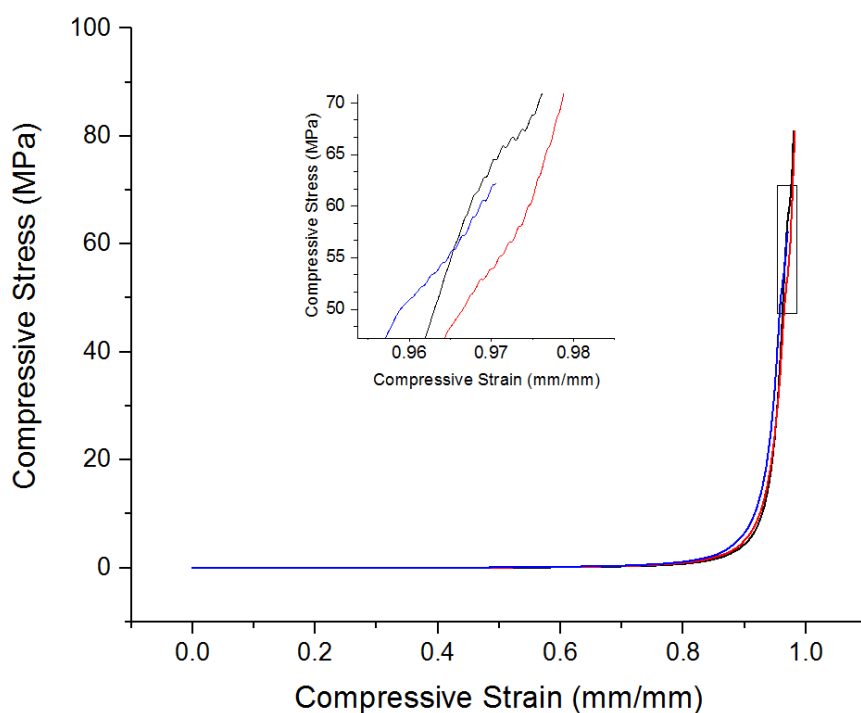
**Figure S7.** Raman spectra recorded for an ArGO film and an ArGO aerogel (indicated). Conditions: ArGO film,  $0.2 \text{ mg mL}^{-1}$  aqueous dispersion GO + agar (agar/GO = 10:1 w/w) +  $\text{NH}_3$  (aq.) (pH = 8-9),  $95 \text{ }^\circ\text{C}$  for 3 h; ArGO gel,  $1.0 \text{ mg mL}^{-1}$  aqueous dispersion GO + agar (agar/GO = 10:1 w/w) +  $\text{NH}_3$  (aq.) (pH = 8-9),  $95 \text{ }^\circ\text{C}$  for 3 h.



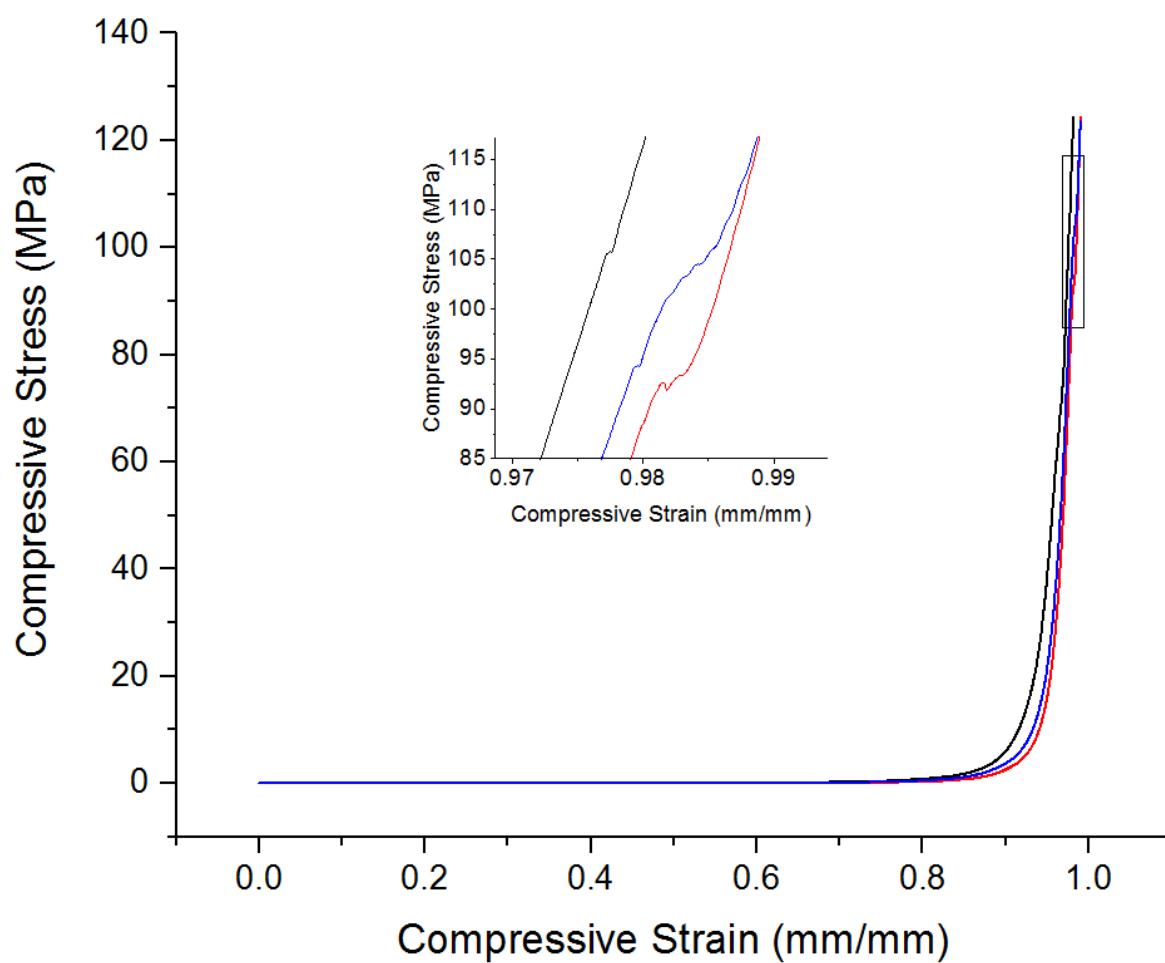
**Figure S8.** FT-IR spectra recorded for GO, agar and ArGO (indicated). Conditions:  $1.0 \text{ mg mL}^{-1}$  aqueous dispersion GO + agar (agar/GO = 10:1 w/w) +  $\text{NH}_3$  (aq.) (pH = 8-9),  $95 \text{ }^\circ\text{C}$  for 3 h.



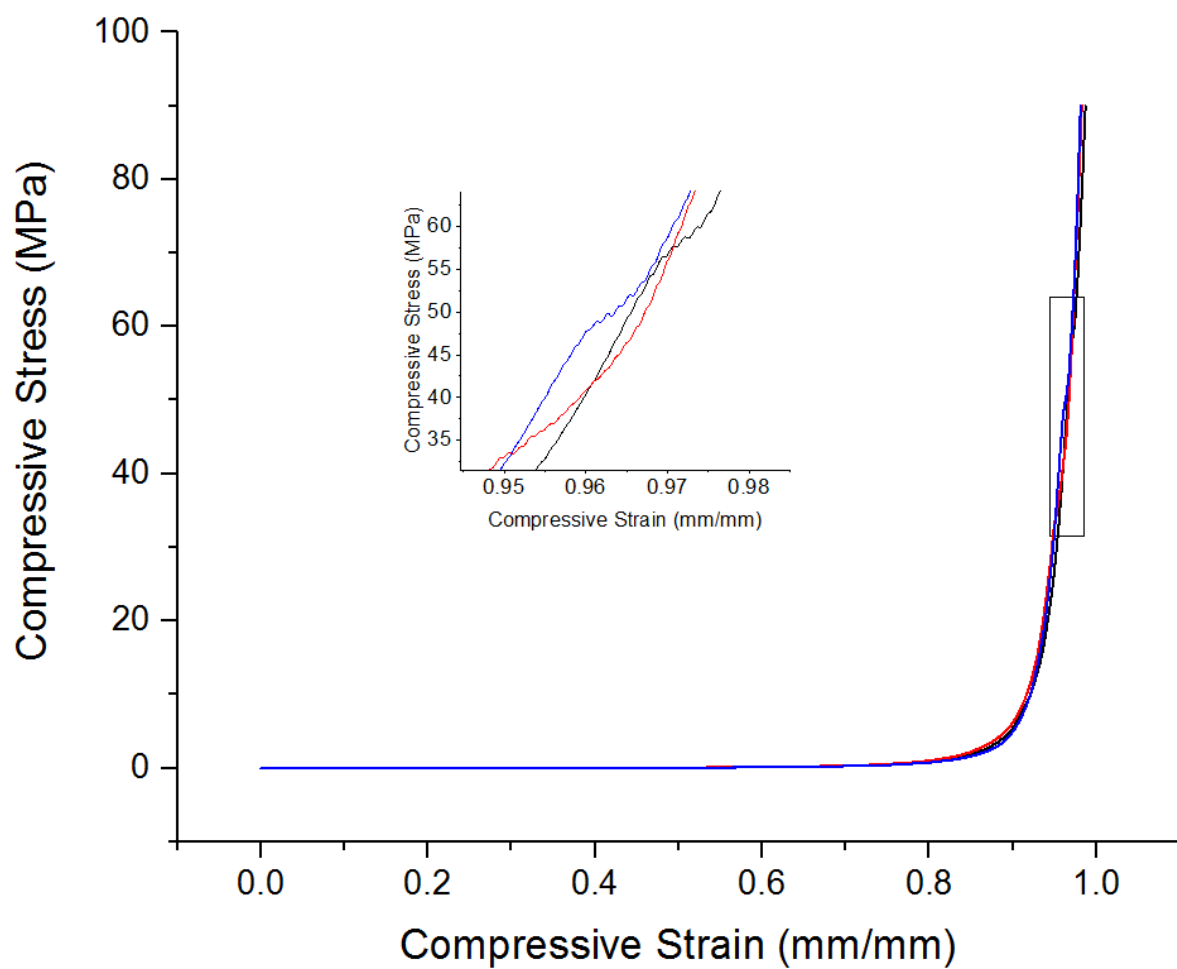
**Figure S9.** FT-IR spectra recorded for PAAm, Agar-PPAm DN gel and ArGO-PAAm DN aerogel (indicated). The gels were freeze-dried for 2 days and then analyzed. Conditions: PAAm, 0.1 mol% MBAA relative to AAm and 1 mol% APS relative to AAm, polymerization time: 7 h at 65 °C; Agar-PAAm DN hydrogel, Agar/AAm = 1: 9 w/w, 0.1 mol% MBAA relative to AAm and 1 mol% APS relative to AAm, polymerization time: 7 h at 65 °C; ArGO-PAAm DN hydrogel, ArGO/AAm = 1: 9 w/w; 0.1 mol% MBAA relative to AAm and 1 mol% APS relative to AAm, polymerization time: 7 h at 65 °C.



**Figure S10.** Stress–strain curves that were recorded for an ArGO-PAAm DN hydrogel. Conditions: ArGO/AAm = 1 : 9 w/w; 0.1 mol% MBAA relative to AAm and 1 mol% APS relative to AAm; polymerization time: 3.5 h at 65 °C; water content: 83 wt%.

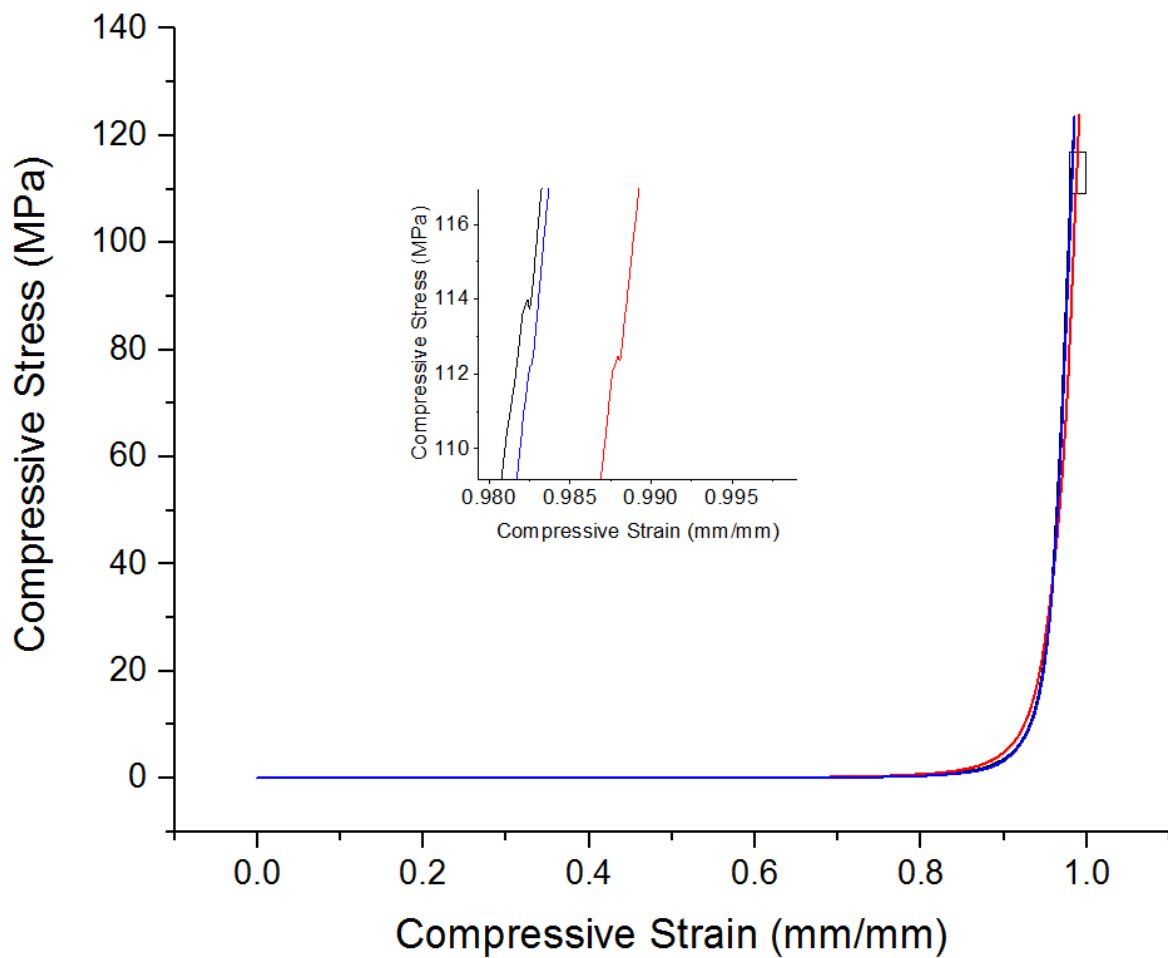


**Figure S11.** Stress–strain curves that were recorded for an ArGO-PAAm DN hydrogel. Conditions: ArGO/AAm = 1 : 9 w/w, 0.1 mol% MBAA relative to AAm and 1 mol% APS relative to AAm, polymerization time: 7 h at 65 °C; water content: 83 wt%.

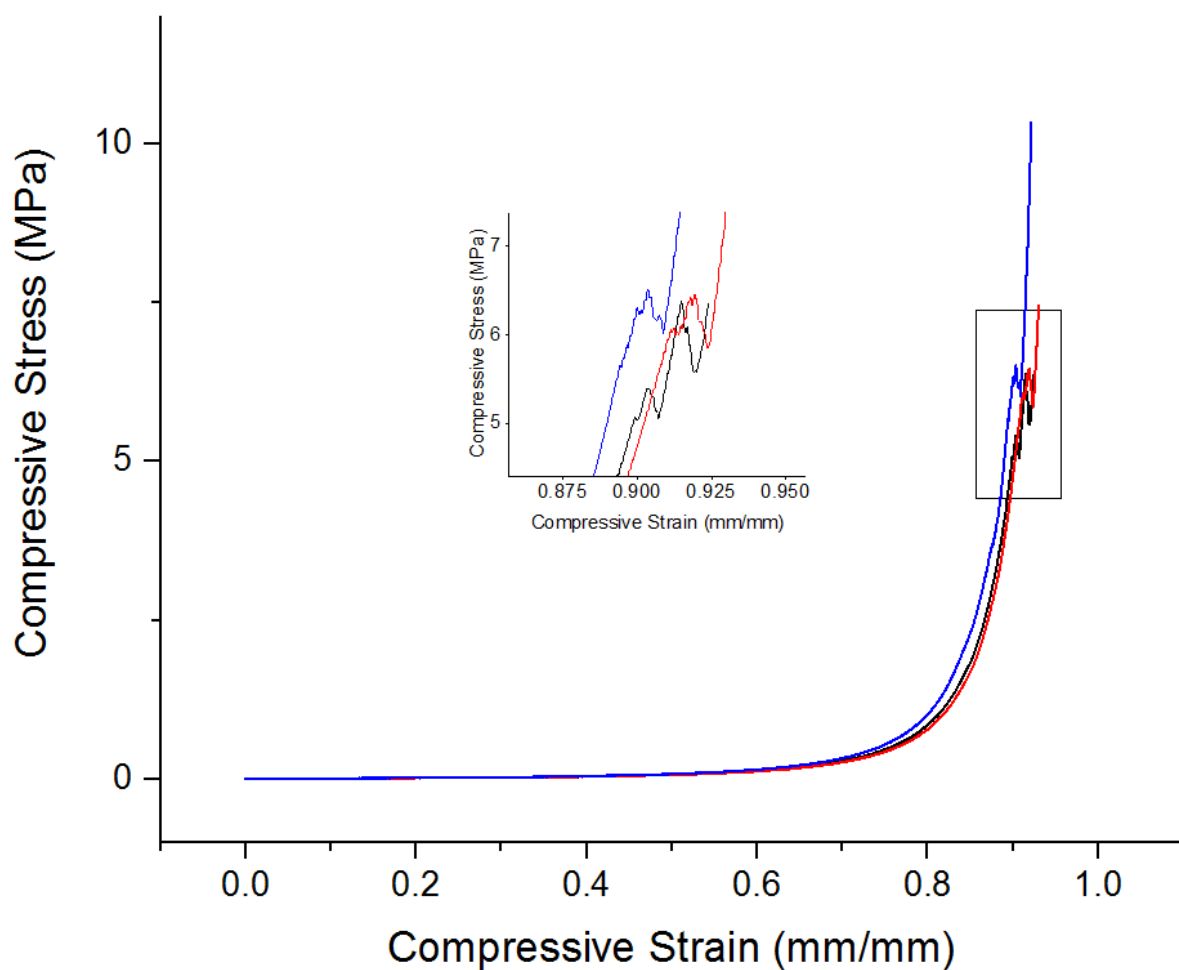


**Figure S12.** Stress–strain curves that were recorded for an ArGO-PAAm DN hydrogel. Conditions: ArGO/AAm = 1 : 9 w/w; 0.1 mol% MBAA relative to AAm and 1 mol% APS relative to AAm; polymerization time: 12 h at 65 °C; water content: 83 wt%.

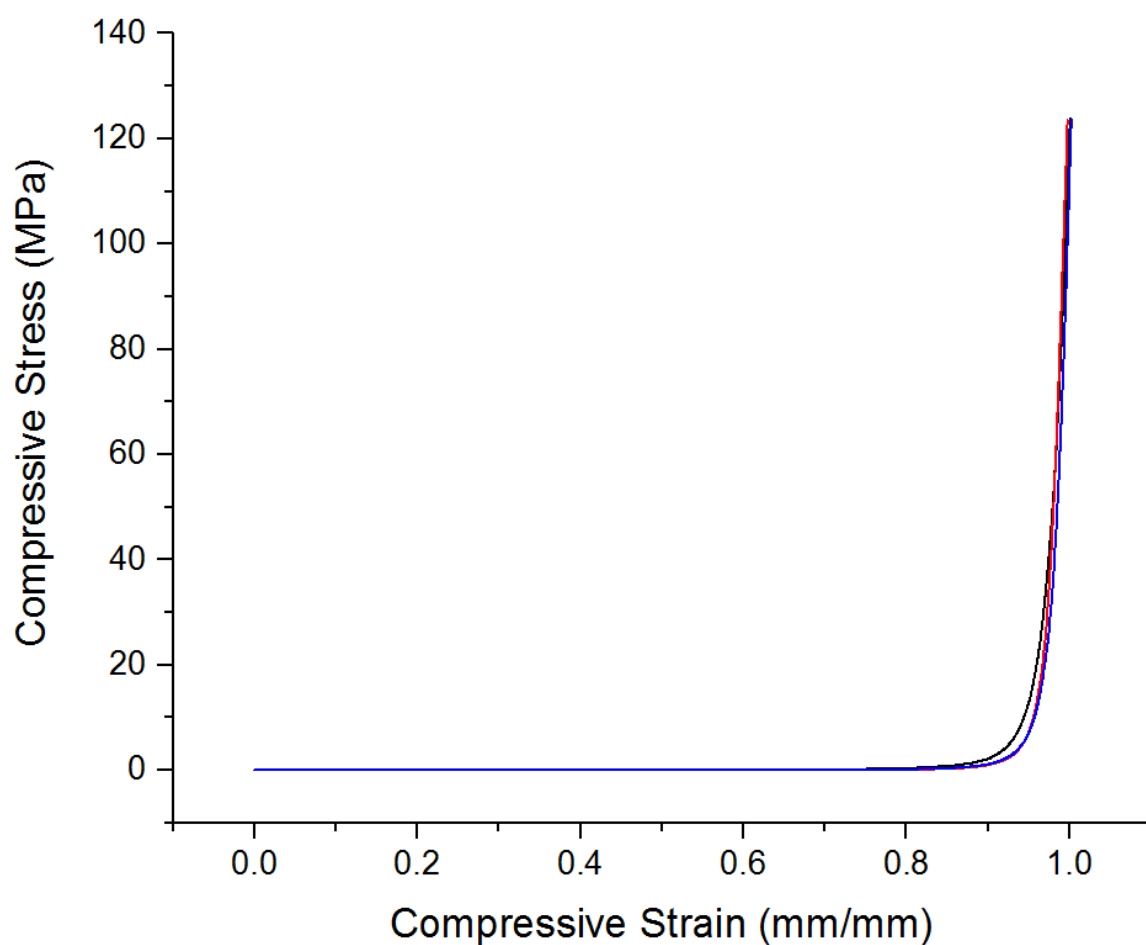




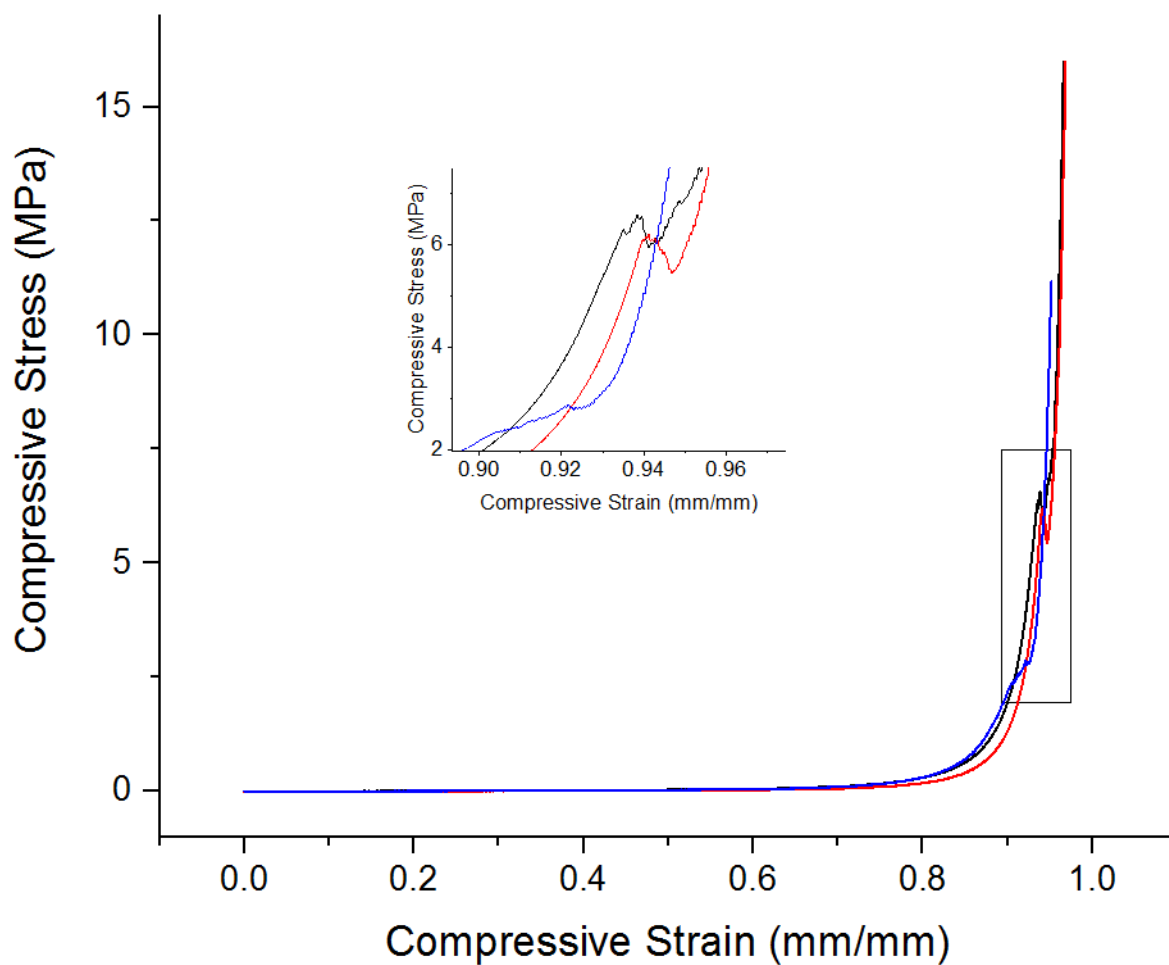
**Figure S13.** Stress–strain curves that were recorded for an ArGO-PAAm DN hydrogel. Conditions: ArGO/AAm = 1 : 9 w/w, 0.06 mol% MBAA relative to AAm and 1 mol% APS relative to AAm, polymerization time: 7 h at 65 °C; water content: 83 wt%.



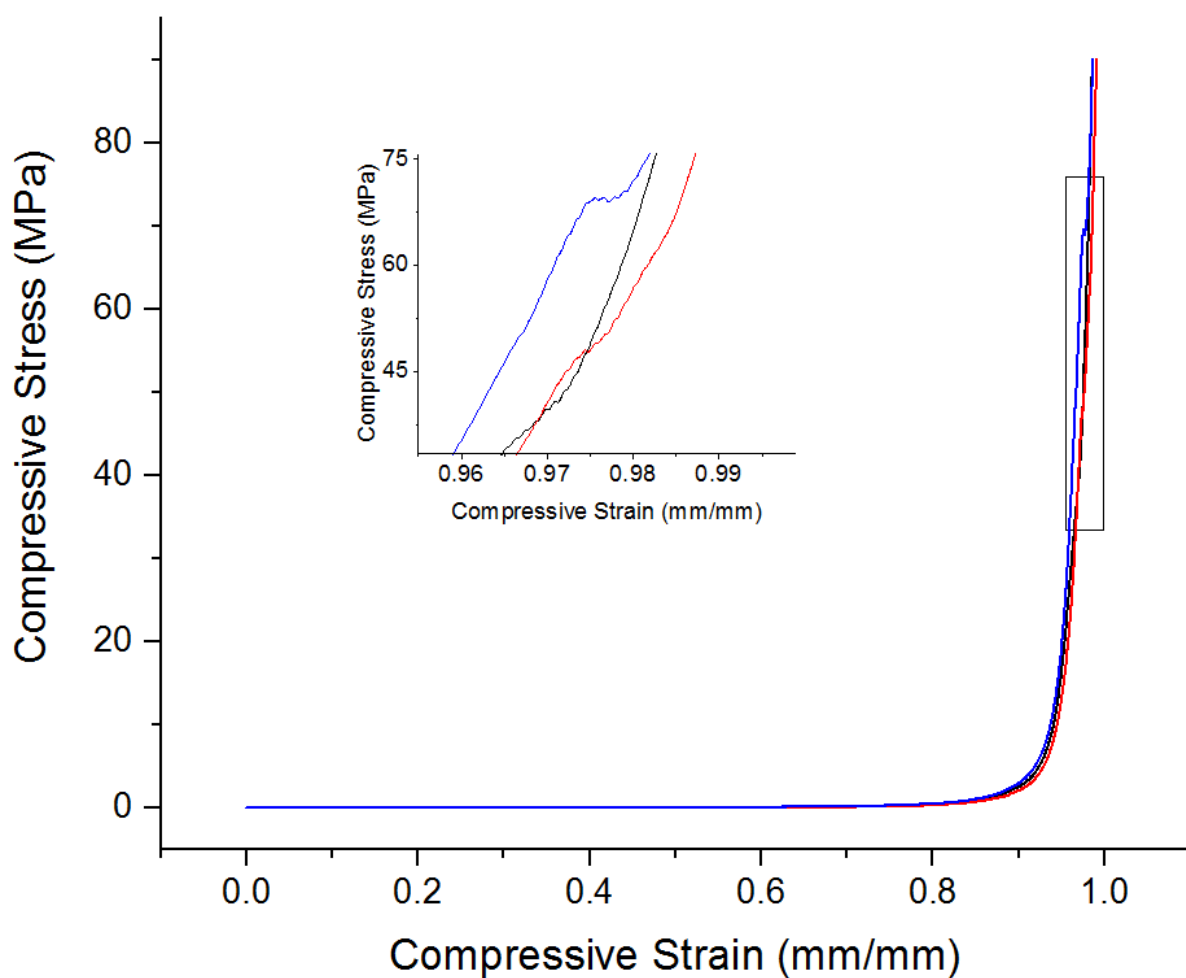
**Figure S14.** Stress–strain curves that were recorded for an ArGO-PAAm DN hydrogel. Conditions: ArGO/AAm = 1 : 9 w/w, 0.025 mol% MBAA relative to AAm and 1 mol% APS relative to AAm, polymerization time: 7 h at 65 °C; water content: 83 wt%.



**Figure S15.** Stress–strain curves that were recorded for an ArGO-PAAm DN hydrogel. Conditions: ArGO/AAm = 1 : 5 w/w, 0.1 mol% MBAA relative to AAm and 1 mol% APS relative to AAm, polymerization time: 7 h at 65 °C; water content: 89 wt%.



**Figure S16.** Stress–strain curves that were recorded for an PAAm SN hydrogel. Conditions: 0.1 mol% MBAA relative to AAm and 1 mol% APS relative to AAm, polymerization time: 7 h at 65 °C; water content: 83 wt%.



**Figure S17.** Stress–strain curves that were recorded for an Agar-PAAm DN hydrogel. Conditions: Agar/AAm = 1 : 9 w/w, 0.1 mol% MBAA relative to AAm and 1 mol% APS relative to AAm, polymerization time: 7 h at 65 °C; water content: 83 wt%.