SUPPORTING INFORMATION

A study of conductive hydrogel composites of pH-responsive microgels and carbon nanotubes

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Table of contents **Page**

I. Figures

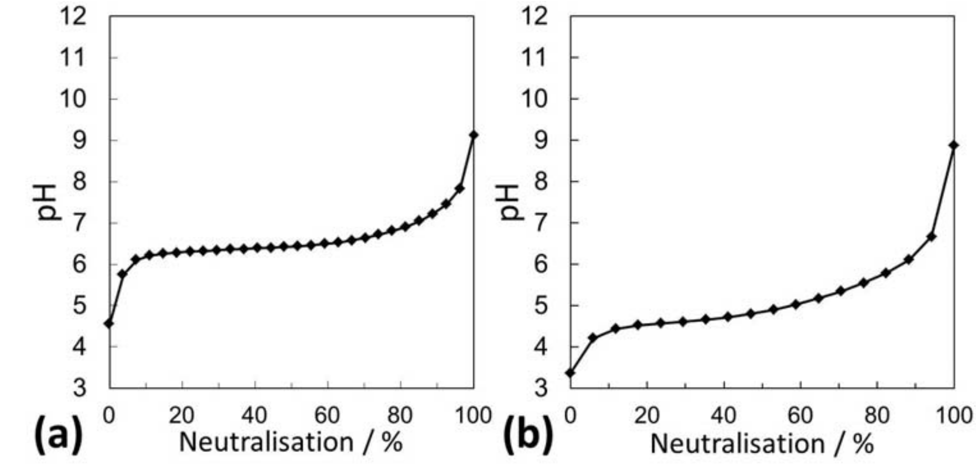

Fig. S1. Potentiometric titration data for MG1 (a) and MG2 (b).

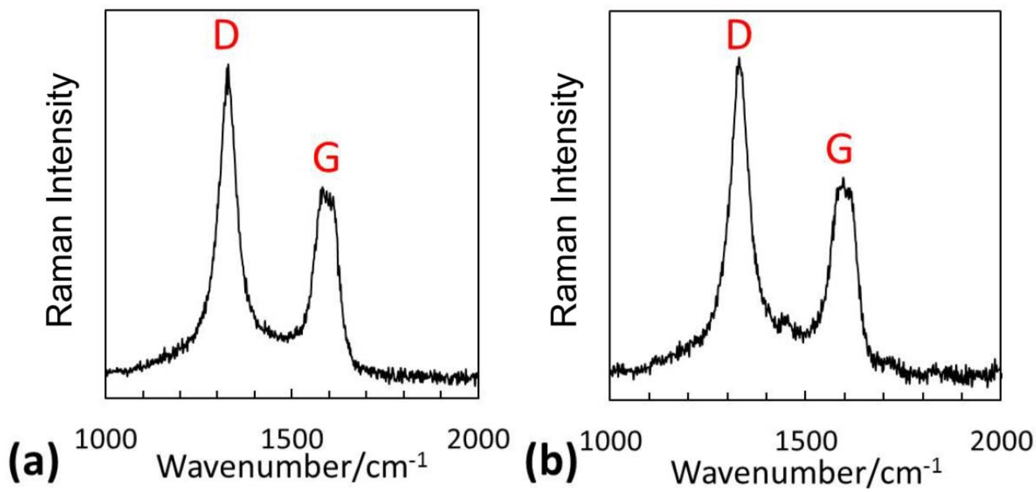

Fig. S2. Raman spectra for as-made and dispersed CNTs. (a) as-supplied CNT and (b) CNTs dispersed using MG1.

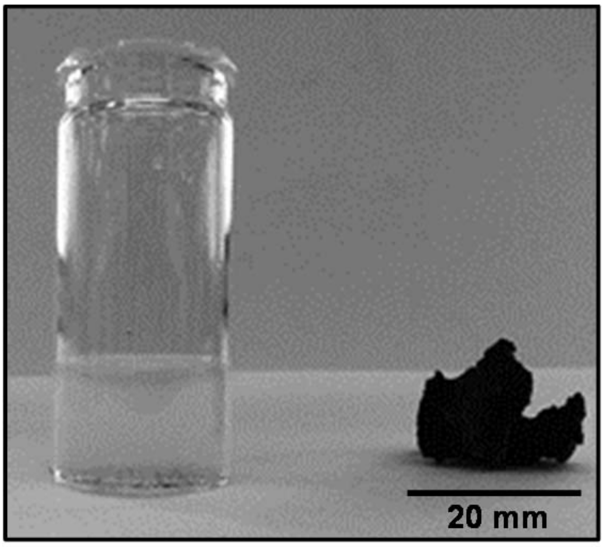

Fig. S3. Colloidal instability of mixed MG1/CNT dispersions at pH 5.0. After mixing, the MG and CNTs formed a macroscopic aggregate.

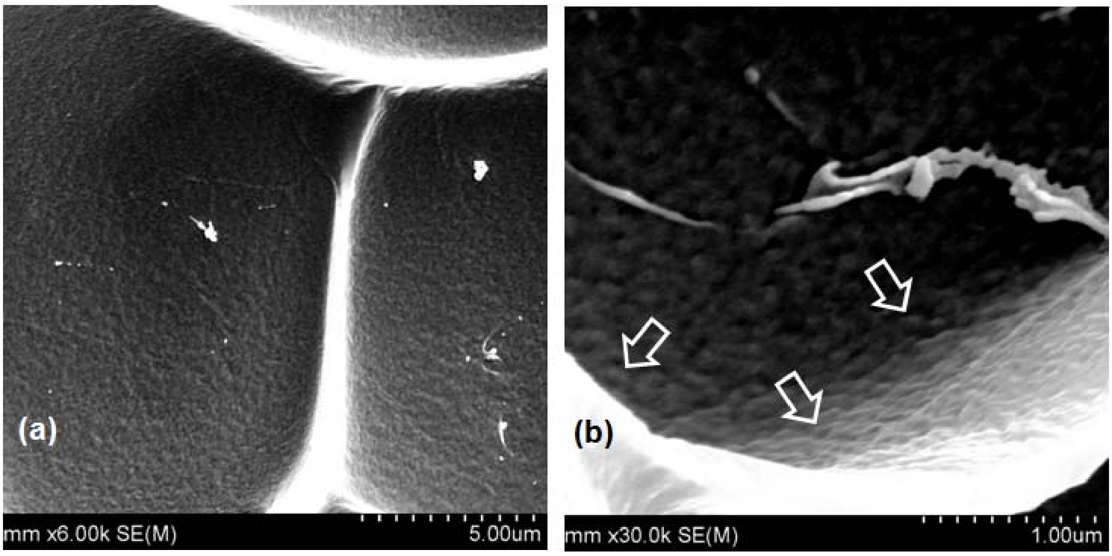

Fig. S4. SEM images showing the morphology of DX MG2/$CNT_{1.0}$ gel composite. Individual MG2 particles can be seen from both images and are highlighted with arrows for (b).

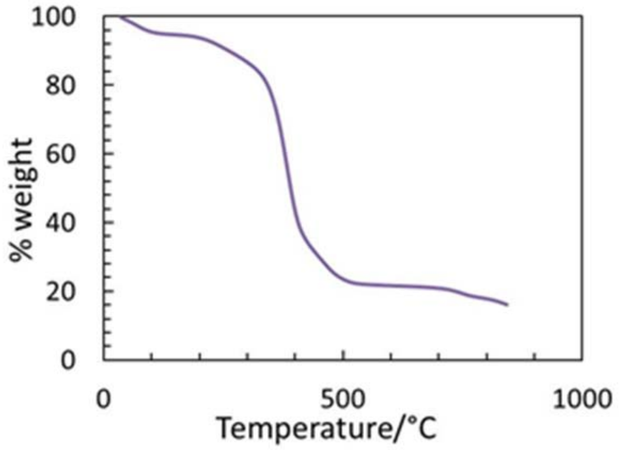

Fig. S5. TGA data for DX MG1/CNT$_{0.5}$. The data were obtained by heating in a N_2 atmosphere at a rate of 10 ^{o}C/ min.

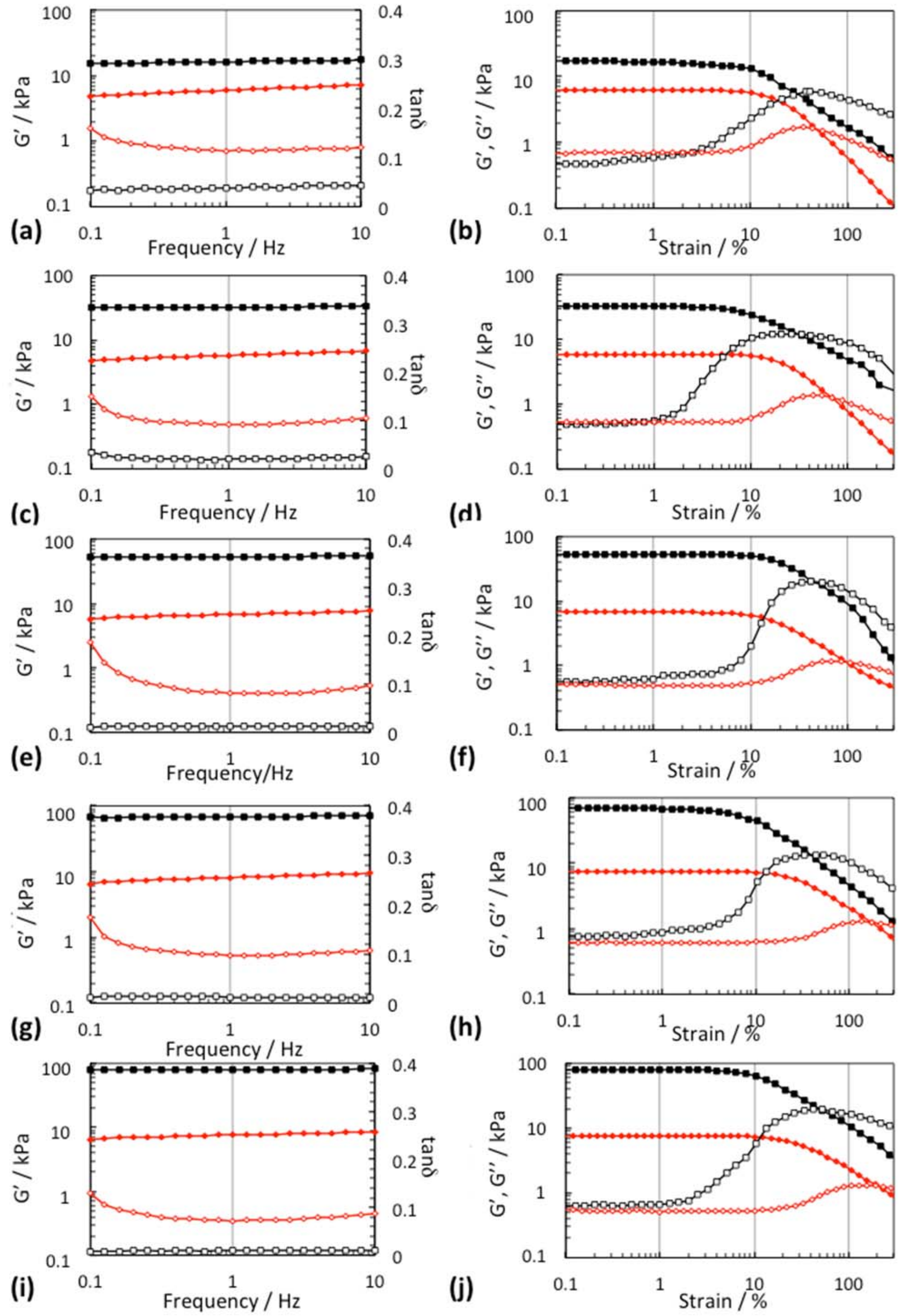

Fig. S6. **Dynamic rheology data for various hydrogel composites.** Black and red symbols apply to DX MG1/CNT and SX MG1/CNT, respectively. Frequency-sweep data are shown in the left hand column. The closed and open symbols are *G'* and tan δ, respectively. Strain-sweep data are shown in in the right hand column. The closed and open symbols are *G'* and *G"*, respectively. The concentrations of CNT used to prepare the composite gels were 0 ((a) and (b)), 0.25 ((c) and (d)), 0.50 ((e) and (f)), 0.75 ((g) and (h)) and 1.0% ((i) and (j)).

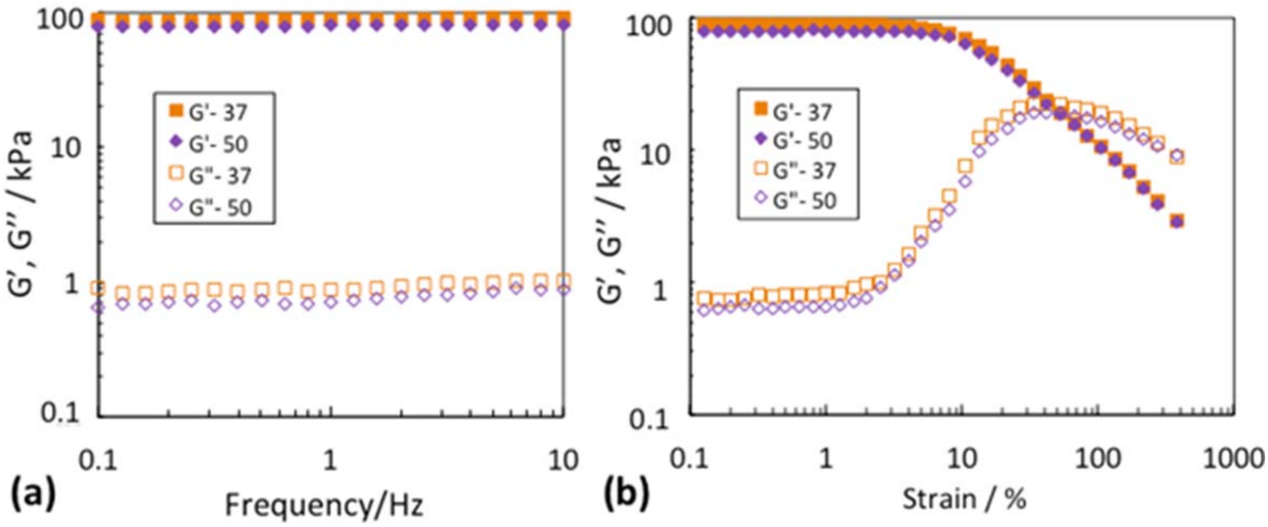

Fig. S7. **Comparison of rheology data for DX MG1/CNT$_{1.0}$ prepared at 37 °C and 50 °C.** (a) Frequency-sweep data. (b) Stain-sweep data. For both figures *G'* and *G"* values are shown as closed and open symbols, respectively. The legends show the temperatures used (^{o} C).

II. Table

Table S1 characterisation data for the microgels

Abbreviation	Mol.% MAA[a]	Mol.% GMA[a]	$d_{n(SEM)}$ [b]/ nm	$d_{h(4)}$ [c]/ nm	$d_{h(10)}$ [c]/ nm	$Q_{MG(10)}$ [d]	pK_a [e]
MG1	38.2	8.0	74 (12)	77	278	47	6.3
MG2	27.4	8.3	111 (17)	121	402	37	4.9

[a] Calculated from potentiometric titration data. [b] Number-average diameters determined from SEM. The number in brackets is the coefficient of variation. [c] The hydrodynamic diameters (d_h) at pH 4.0 and 10.0. [d] Swelling ratio calculated at pH 10.0 using $Q_{MG} = (d_{h(10)} / d_{h(4)})^3$. [e] The apparent p$K_a$ values were determined from potentiometric titration data.

III. Derivation of equation for maximum average fractional coverage of MGs by CNTs

A simple derivation is given for an equation for the maximum average fractional coverage of MG particles by CNTs, i.e., $\theta_{MG(max)}$. The collapsed and swollen diameters for the MG particles are $D_{MG(coll)}$ and D_{MG}, respectively. The MG particles are present in the gel composite with a total dry volume fraction of $\phi_{MG(T)}$. The volume swelling ratio for the MG particles is Q_{MG} (= $(D_{MG}/D_{MG(coll)})^3$). The average length and diameter of the CNTs are L_{CNT} and D_{CNT}, respectively, and the CNTs are present in the composite with a volume fraction of $\phi_{CNT(T)}$. Because of their very high aspect ratios CNT end effects are ignored in the following. The steps for deriving an expression for $\theta_{MG(max)}$ are to obtain expressions for the total surface areas of the MG particles ($A_{MG(T)}$) and the CNTs ($A_{CNT(T)}$) and then to use those expression to obtain the final equation for $\theta_{MG(max)}$.

An expression for $A_{MG(T)}$ can be obtained using the product of the number of MG particles present in the gel (N_{MG}) and the surface area for each MG particle (A_{MG}). An expression for N_{MG} can be found from the ratio of total dry volume of MG particles to the volume of an individual MG particle in the collapsed state.

$$N_{MG} = \frac{6\phi_{MG(T)}V_T}{\pi D_{MG(coll)}^3} \qquad \text{(S1)}$$

For equation S1, V_T is the total volume of the composite gel. The following expression for $A_{MG(T)}$ can be found using equation S1, A_{MG} (= πD_{MG}^2) and the equation for Q_{MG} (above).

$$A_{MG(T)} = \frac{6\phi_{MG(T)}V_T Q_{MG}^{2/3}}{D_{MG(coll)}} \qquad \text{(S2)}$$

An equivalent approach was used for the CNTs to that described above which gave the following.

$$A_{CNT(T)} = \frac{4\phi_{CNT(T)}V_T}{D_{CNT}} \qquad \text{(S3)}$$

From the ratio of equations S2 and S3 we obtain the final equation:

$$\theta_{MG(max)} = \frac{2\phi_{CNT(T)}D_{MG(coll)}}{3\phi_{MG(T)}D_{CNT}Q_{MG}^{2/3}} \qquad \text{(S4)}$$