SUPPLEMENTARY INFORMATION

Doubly crosslinked microgel / polyelectrolyte complexes: three simple methods to tune and improve gel mechanical properties

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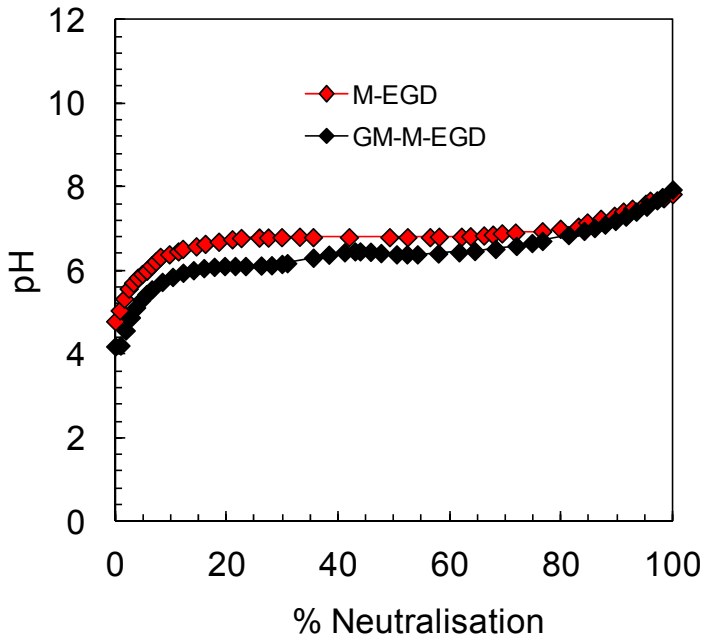

Fig. S1 Potentiometric titration data for GM-M-EGD and M-EGD MGs.

^{1}H NMR spectra analysis

Each of the PAH*w*k-*x*AMAm spectra (Fig. S2) show polyallylamine (PAH) signals (*a*, *b* and *c* protons) as well as new peaks due to the allyl methacrylate (AMAm) units. The latter are evidenced by the vinyl protons (*e*) as also the methylenic protons adjacent to the amide group (*c'*). The increase in the relative sizes of these peaks shows that vinyl functionalisation was successful and increased in the order PAH15k-11AMAm < PAH65k-35AMAm < PAH15k-55AMAm. To determine values of *x* for PAH*w*k-*x*AMA we used the ratio of the integrated areas for the vinyl (*e*) protons (*A(e)*) and those responsible for the signals at chemical shift (δ) values less than 2 ppm; that is the protons *a*, *b* and *d*. The equation used to determine the values for *x* was as follows.

$$x = \frac{3}{2R-3} \qquad \text{(S1)}$$

where $R = A(a,b,d) / A(e)$ and $A(a,b,d)$ is the sum of the areas for the *a*, *b* and *d* protons.

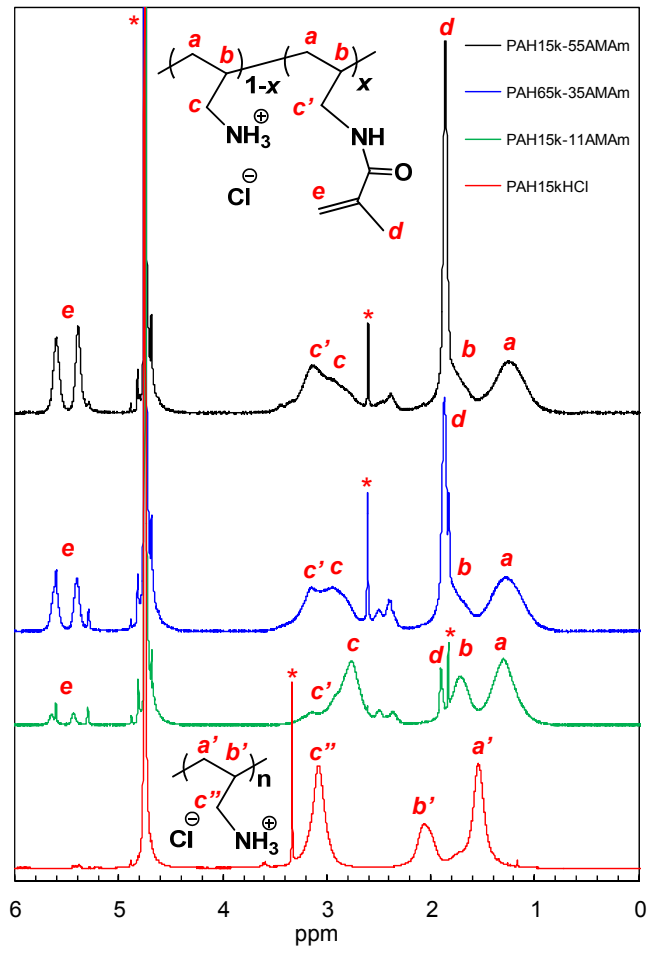

Fig. S2. ^{1}H NMR spectra for polymers obtained using D_2O. Solvent peaks are indicated with asterisks.

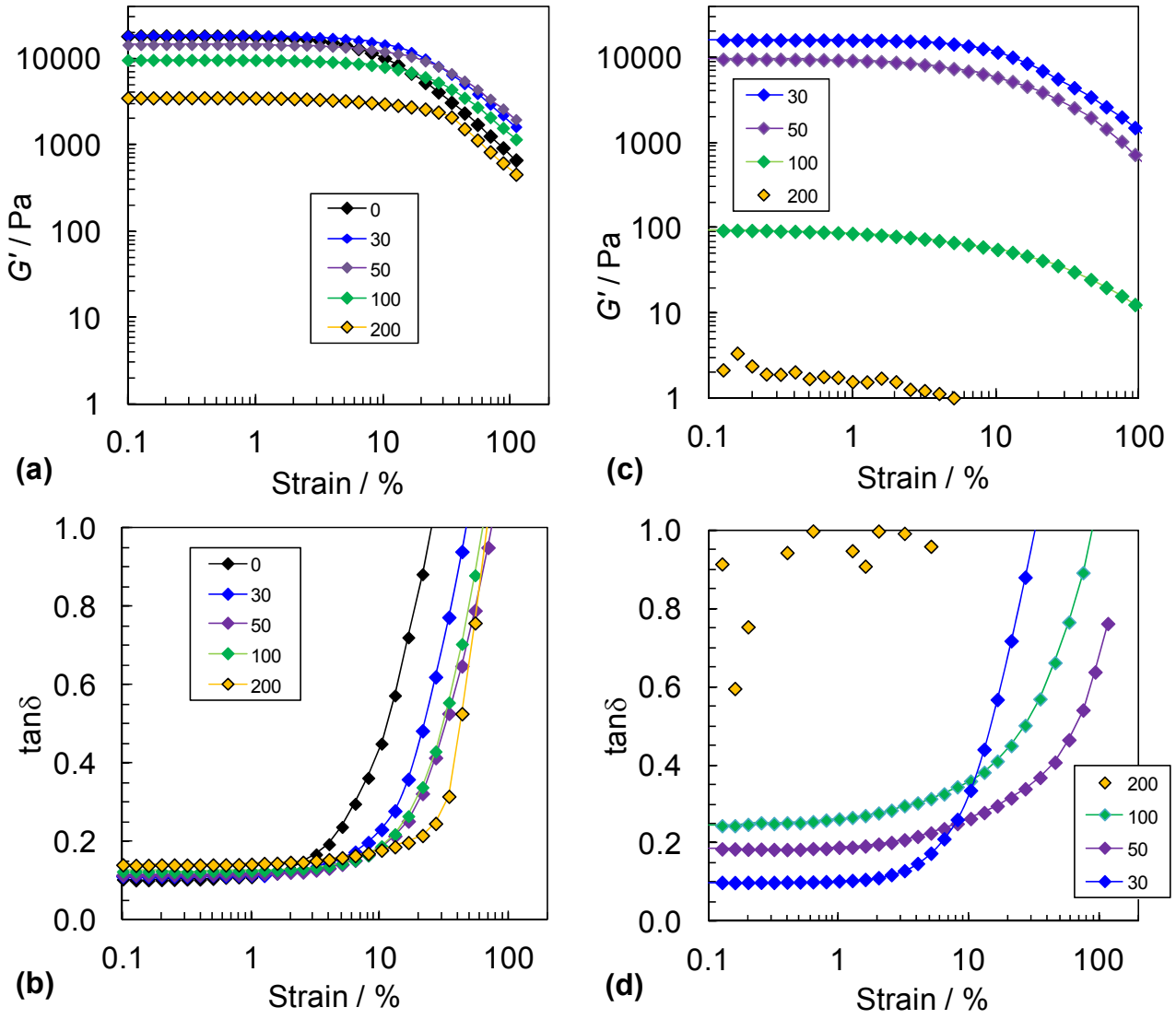

Fig. S3. Strain-sweep dynamic rheology data for DX MG / PAH15k-55AMAm ((a) and (b)) and DX MG / PAH15k ((c) and (d)) obtained at different MR values (shown as mg / g in legends). The data for MR = 0 in (a) and (b) correspond to DX MG.

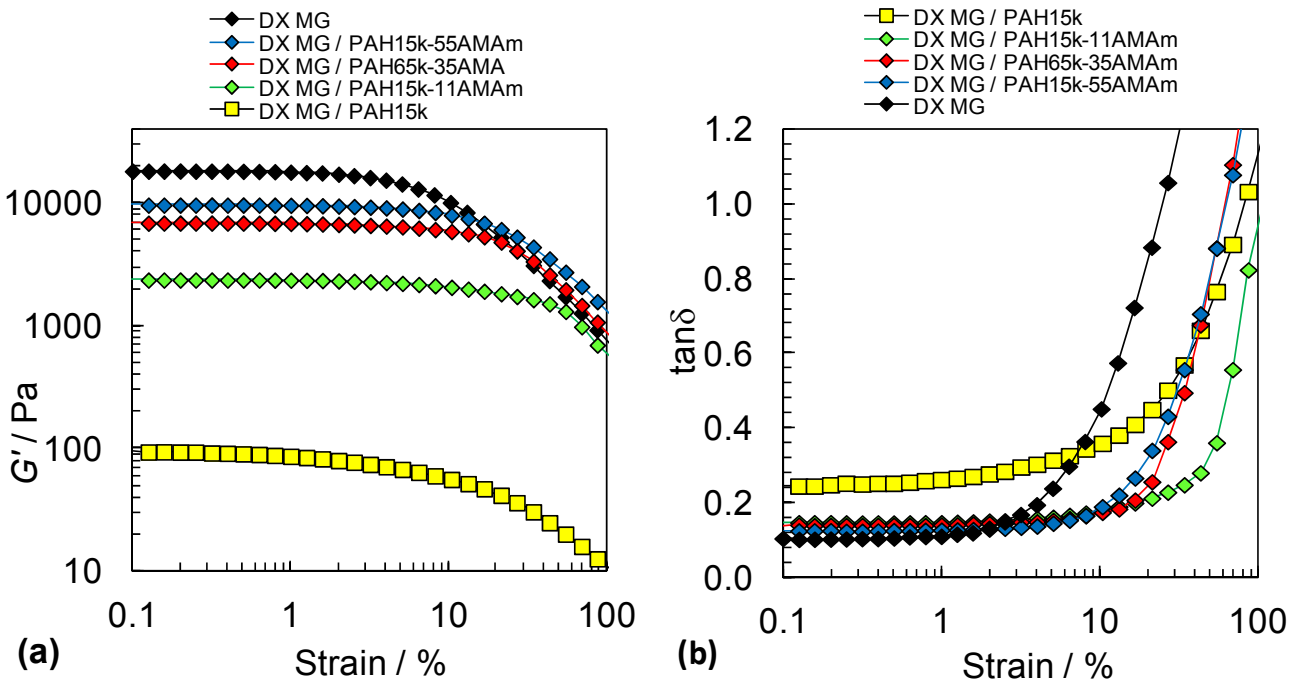

Fig. S4. Strain-sweep dynamic rheology data for DX / PAH*w*k-*x*AMAm gels. (a) and (b) show the variations of *G'* and tanδ, respectively. Note that MR = 100 mg / g.

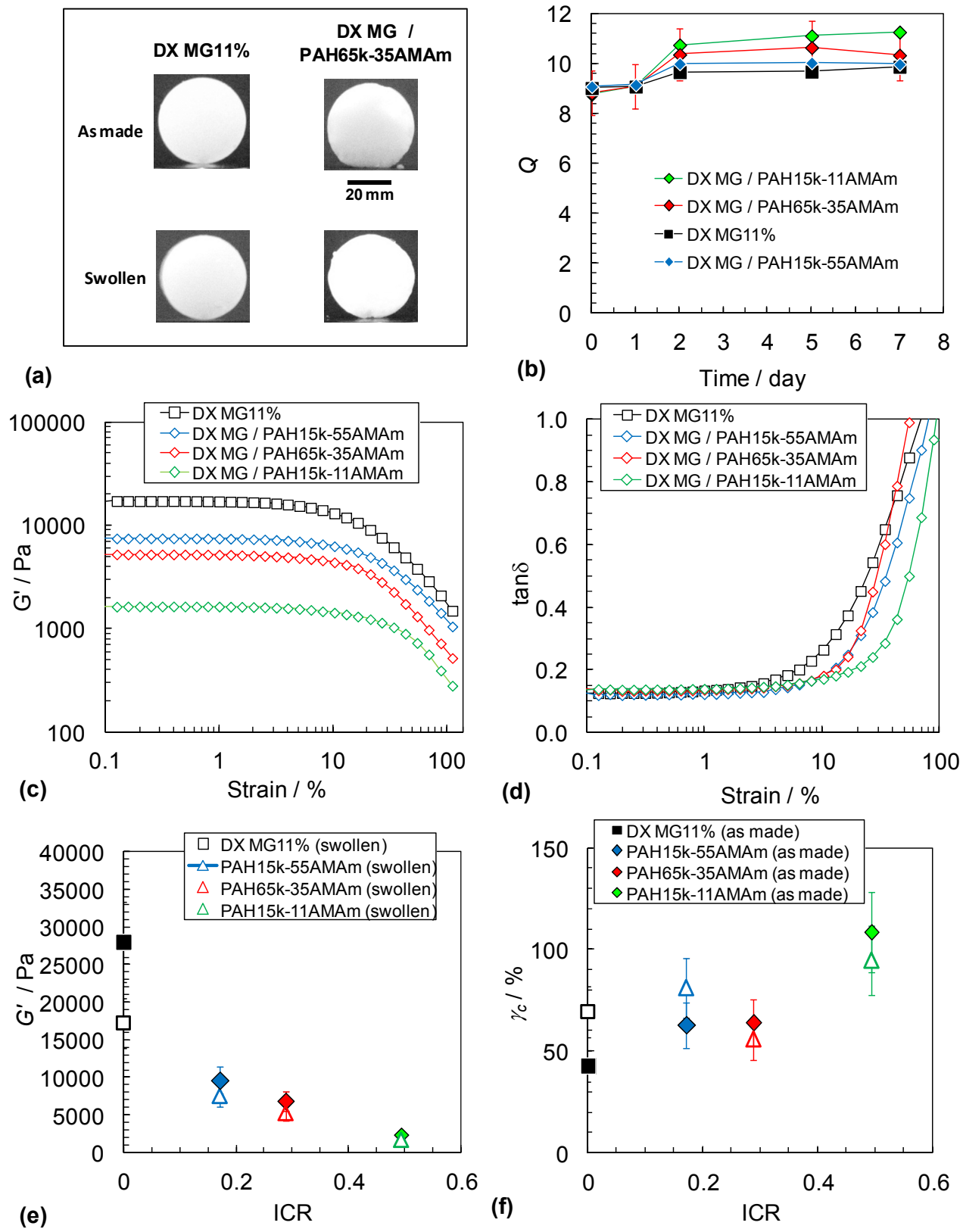

Fig. S5. Swelling data for DX MG / PAH15k-*x*AMAm gels. The gels were placed in pH = 7.5 buffer for 7 days. (a) Shows selected representative digital photographs for the discs before and after swelling. (b) Shows swelling ratios as a function of time. (c) and (d) show strain-sweep data for the swollen discs. (e) and (f) show *G'* data and γ_c data, respectively. For (e) and (f) the same legends apply to both graphs. Note that the samples were prepared at MR = 100 mg /g.

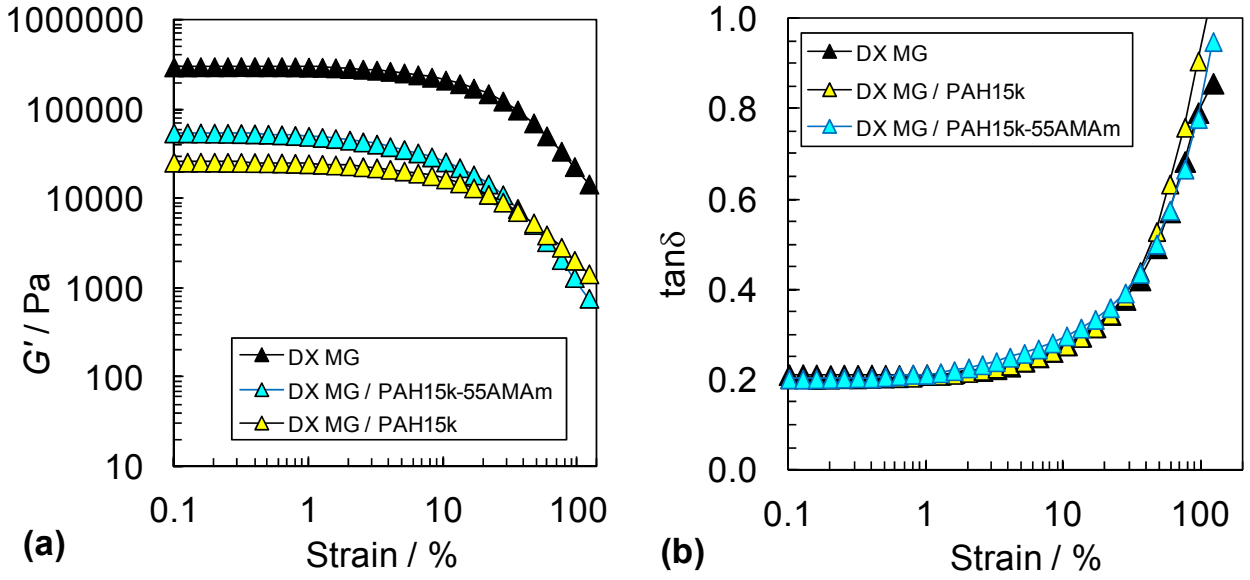

Fig. S6. Strain sweep data for DX MG / PEC and DX MGs prepared in aqueous 1 M NaCl solution. The samples were prepared using MR = 100 mg / g.

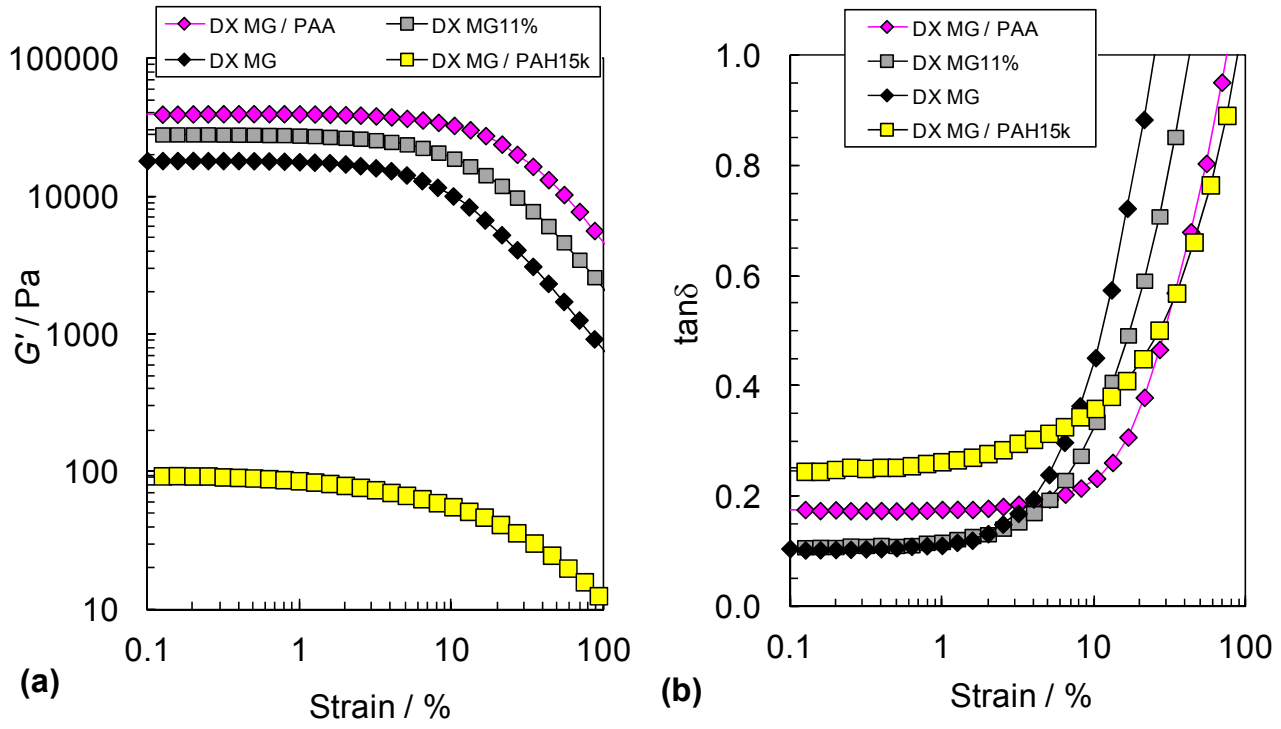

Fig. S7. Strain sweep data for DX MG / polyelectrolyte and DX MGs. The wt.% of DX MG used is indicated. For the gels MR = 100 mg / g. See text.