

Supplementary Information

Fig. S1. Potentiometric titration data for the microgels. pH and pK_a data are shown for M-EGD, GM-M-EGD and GM(H)-M-EGD in (a) and (b), respectively. Data for E-BDD and GM-E-BDD are shown in (c) and (d).



Fig. S2 Scanning electron micrographs for M-EGD (top left), GM-M-EGD (top right), E-BDD (bottom left) and GM-E-BDD (bottom right).



Fig. S3. Frequency sweep dynamic rheological data for DX GM-M-EGD microgels prepared and measured at different pH values (shown in legend). The value for ϕ_p was 0.10.



Fig. S4. Effect of ϕ_p of GM-M-EGD used during double crosslinking on (a) *G*' and (b) tan δ as a function of strain. The values for ϕ_p used during preparation are shown in the legend. The pH used to prepare the DX microgels was 7.8.



Fig. S5. Swelling ratios for (a) DX GM-M-EGD and DX GM(H)-M-EGD microgels as well as (b) DX GM-E-BDD microgel as a function of time measured at different pH values. The lines are guides to the eye.

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Fig. S6 Strain amplitude data for DX GM-M-EGD and DX GM(H)-M-EGD ((a) and (b)) as well as DX GM-E-BDD ((c) and (d)) microgels after swelling at a range of pH values for 8 days. The DX microgels were prepared at pH = 7.8.