Electronic Supplementary Material (ESI) for Soft Matter.

Electronic Supplementary Information

Magnetic response of gelatin ferrogels across the sol-gel transition: Influence of high energy crosslinking on thermal stability

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2.1 Ferrogel Fabrication

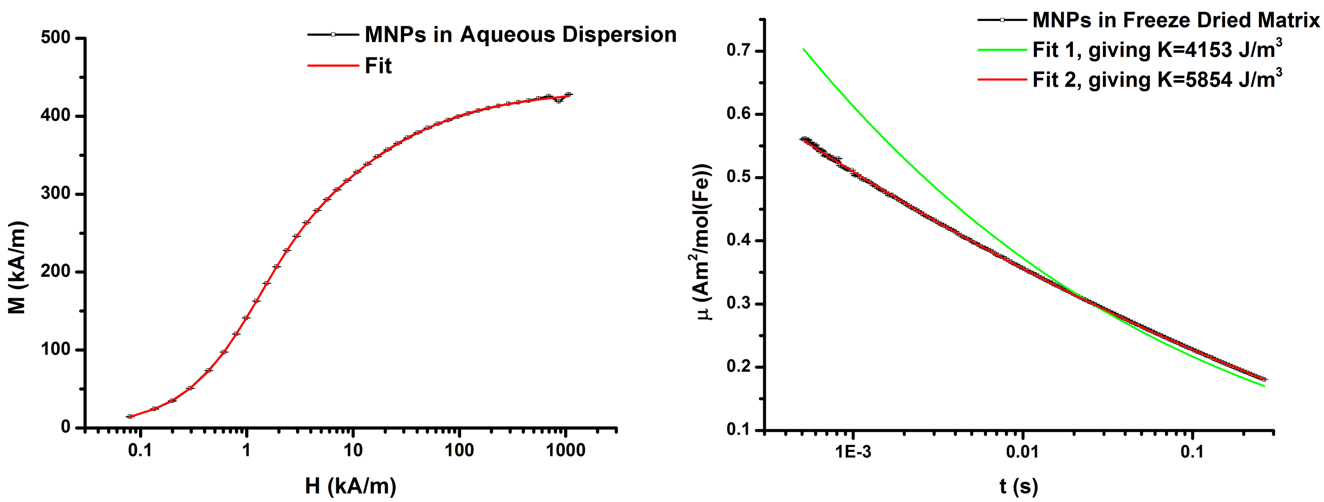

Fig. S7 a) M(H) data of an aqueous dispersion of HK111 MNPs and the MSM[18] fit, yielding a saturation magnetization of $M_s = (432500 \pm 11000)$ A/m, diameter of the mean magnetic volume $d_V = (27.4 \pm 1)$ nm and dispersion parameter $\sigma = 0.1327 \pm 0.04$.
b) MRX data of the immobilized MNPs (freeze dried sample). MSM fits were performed using (Fit 1) the parameters from the M(H) data, yielding K = 4153 J/m and (Fit 2) using the parameters of the M(H) results as constraints with the range according to the determined uncertainties, yielding a saturation magnetization of $M_s = (419320 \pm 11000)$ A/m, diameter of mean magnetic volume $d_V = (24.2 \pm 1)$ nm, dispersion parameter of $\sigma = 0.212 \pm 0.013$ and K = 5854 J/m.

3.2 Effect of chemical crosslinking on MNP distribution

Tab. S1 Percent of MNPs released during irradiation with increasing dose, as determined from magnetic analysis of the liquid byproduct. Significantly less than 1% of the particle content was lost. Slightly larger values measured for the 20 kGy sample could be due to accidental collection of a small sample fragment.

Dose (kGy)	Particles released (%)	Error (%)
10	0.03	0.01
20	0.61	0.14
40	0.10	0.03
100	0.04	0.01

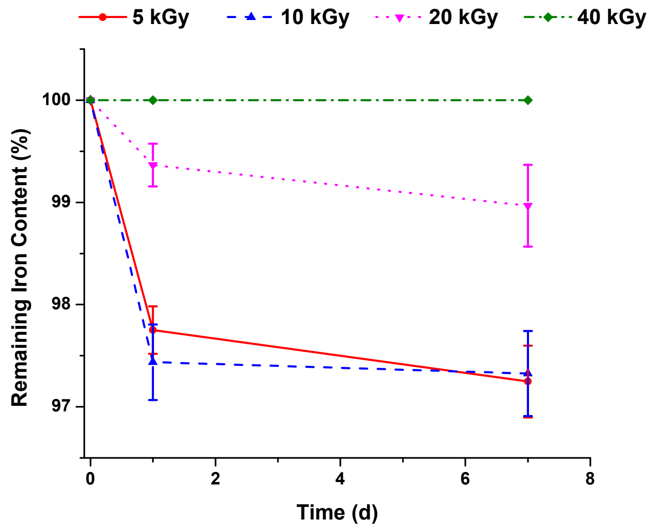

Fig. S8 Iron content remaining in crosslinked ferrogels during incubation in SBF with time. Samples crosslinked with 100 kGy are not shown since the losses were negligible, as in the 40 kGy sample.

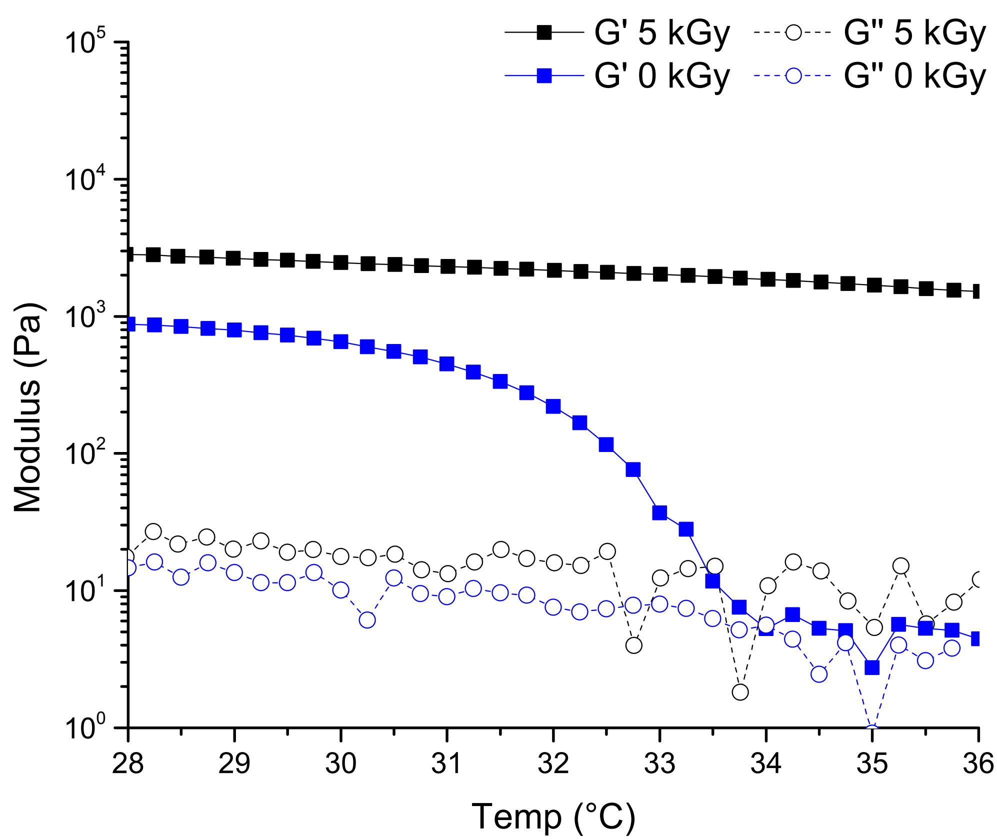

Fig. S9 Macroscopic shear rheology measurements showing the storage (G') and loss (G") modulus for the unirradiated and 5 kGy irradiated 4 wt% gelatin ferrogels across the sol-gel regime.

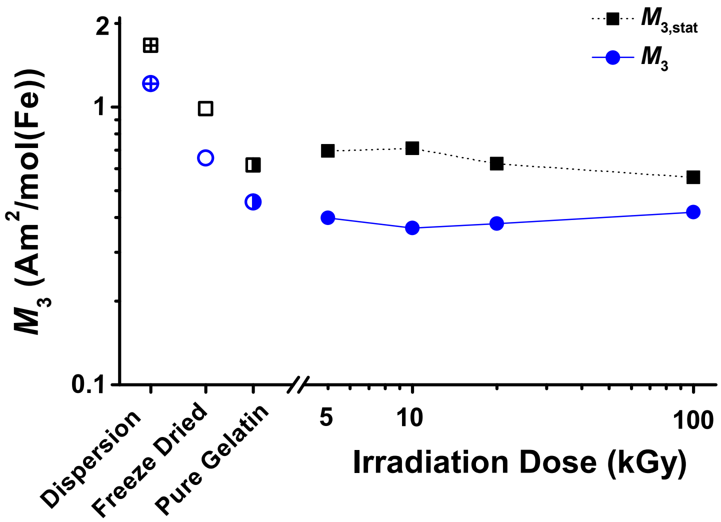

Fig. S10 Magnetic measurements for particles in aqueous dispersion, freeze dried, 4 wt% uncrosslinked gelatin and gelatin with increasing irradiation doses are given. Slight variations in the measured M_3 and $M_{3,stat}$ values for irradiated gelatin samples are not significant, as the samples were prepared outside of the measurement capsules and cut to fit. The iron content was determined by the sample mass. Therefore, a larger error was expected. Furthermore, MPS spectra compared before and after irradiation with 40 kGy revealed no considerable differences in the magnetic response.

3.3 Magnetically-observed thermal response of the physically crosslinked ferrogels

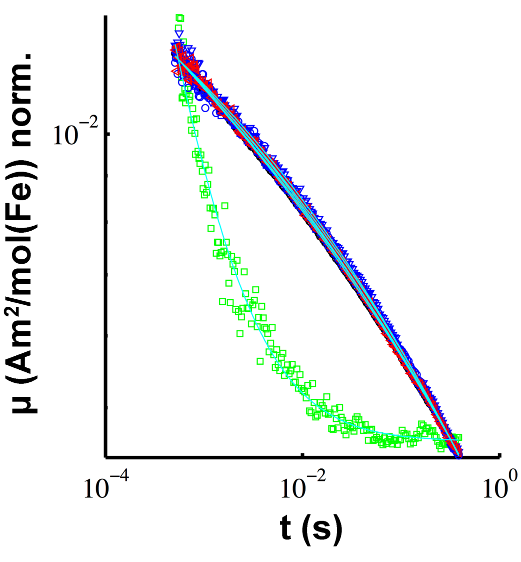

Fig. S11 Normalized MRX spectra for MNPs in aqueous solution (green squares) compared to in freeze dried and gelatin matrices. Spectra for MNPs in freeze dried and gelatin matrices overlap and are not easily distinguished. Samples include freeze dried (black circles) as well as 1.9 wt% (blue squares), 4.5 wt% (blue circles), 8.3 wt% (blue triangles) and 14 wt% gelatin (red triangles). Light blue lines show fits, only distinguishable between the aqueous and freeze dried/gelatin samples.

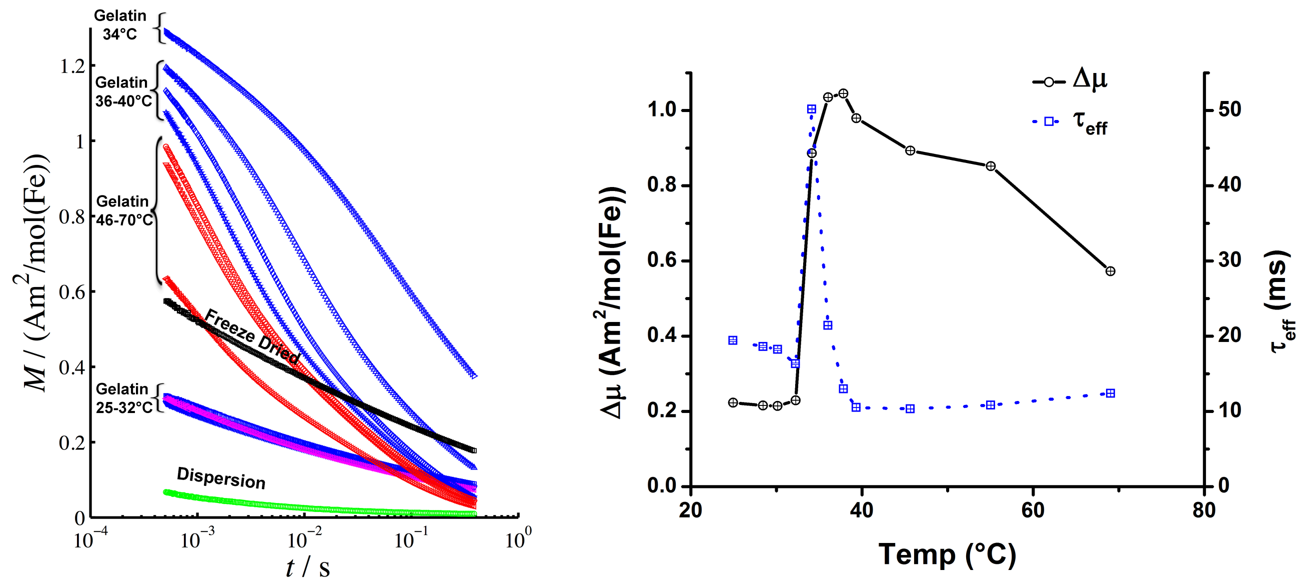

Fig. S12 a) MRX spectra are given for MNPs in aqueous dispersion and a freeze dried matrix, plus in 4 wt% gelatin matrices heated from 25 to 70°C. Spectra for gelatin increase in initial μ values with increasing temperature, peak at 34°C and subsequently decrease with temperature. b) The change in the magnetic moment μ is given for MNPs in gelatin as a function of temperature, along with the effective relaxation times. After the sol-gel transition, $\Delta\mu$ is much larger and decreases at higher temperatures, corresponding to the jump observed in relaxation times.

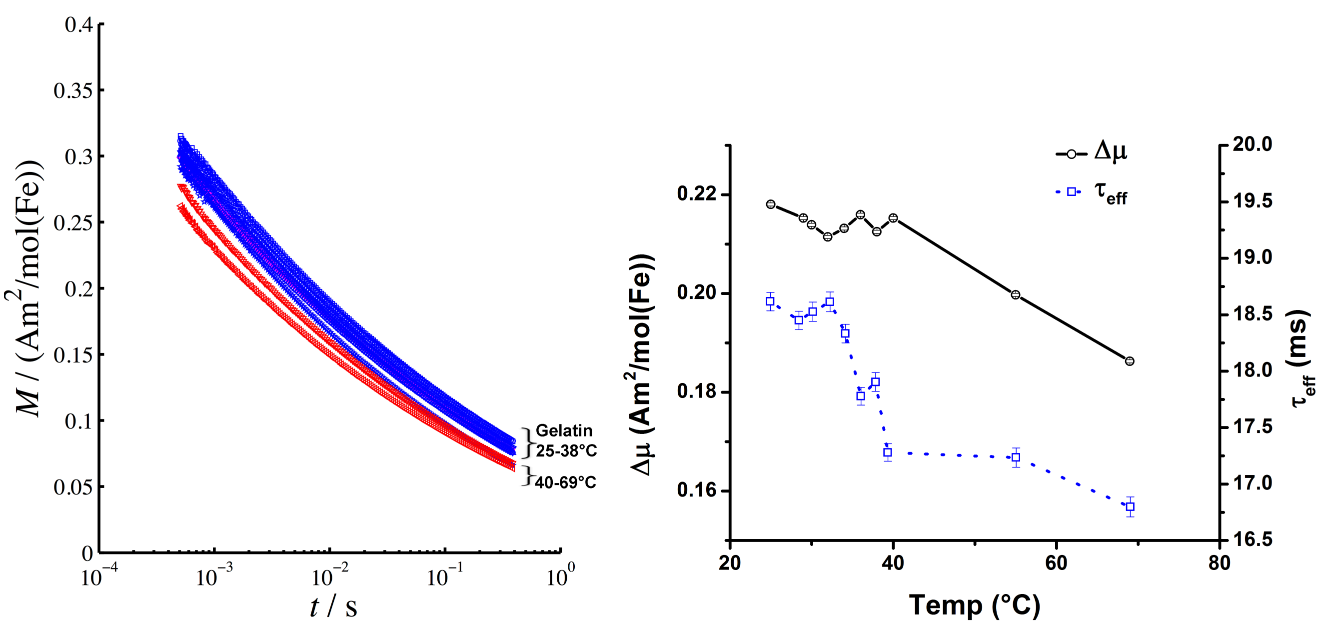

Fig. S13 a) MRX spectra for MNPs in 4 wt% gelatin heated from 25 to 69°C. Generally, the initial moment decreased with increasing temperature. b) The change in the moment observed across the measurement period is highly stable compared to the changes observed in the uncrosslinked gelatin matrices, shown with the effective relaxation times. However, at higher temperatures, a slight decrease is still observed.