Supplementary Information (SI) for Soft Matter.

Journal Name

ARTICLE TYPE

Cite this: DOI: 00.0000/xxxxxxxxxx

Electronic Supplementary Information for "The Impact of Cross-Linker Distribution on Magnetic Nanogels: Encapsulation, Transport and Controlled Release of the Tracer."

Ivan S. Novikau,*[a] Ekaterina V. Novak,[b] and Sofia S. Kantorovich[a,c]

Received Date
Accepted Date

DOI: 00.0000/xxxxxxxxxx

-

1 Analysis of Bond Length Distribuitions

The energy of a bond is proportional to its length, l_{bond}. Hence, in Fig. 1**a** (right) we present the density distribution of normalized bond lengths within a specific morphology, $\rho(\overline{l_{bond}})$, where $\overline{l_{bond}} = (l_{bond} - l_{bond,min})/(l_{bond,max} - l_{bond,min})$. This 'min-max' scaling is chosen to highlight the presence of highly stretched cross-linkers within nonuniform morphologies. Non-rescaled distributions, $\rho(l_{bond})$, are provided in Fig. 1**a** (left). A comparison between the plots in Fig. 1**a** reveals that rescaling only affects cross-linkers, meaning that the length of polymer backbone stretchable springs remains consistent across all morphologies. Additionally, the distribution of average bond length over a small shell distance r from the center of mass of the gel is presented in Fig. 1**b**. We once again observe that polymer l_{bond} remain constant throughout the gel's volume, while cross-linkers exhibit fluctuations. For instance, there is a peak for the gaussian morphology at the center of the gel. By correlating average l_{bond} for different morphologies with bond stretching energies, E_{bonded}, shown in Fig. 1**c**, we can infer that due to excluded volume interactions, it becomes challenging for cross-linkers to pack polymers into confined spaces, resulting in the stretching of cross-linkers.

[a] Faculty of Physics, University of Vienna, Kolingasse 14-16, Vienna 1090, Austria E-mail: ivan.novikau@univie.ac.at
[b] Ural Federal University, Lenin Av. 51, Ekaterinburg 620000, Russian Federation
[c] Research Platform MMM, University of Vienna, Oskar-Morgenstern-Platz 1, Vienna 1090, Austria

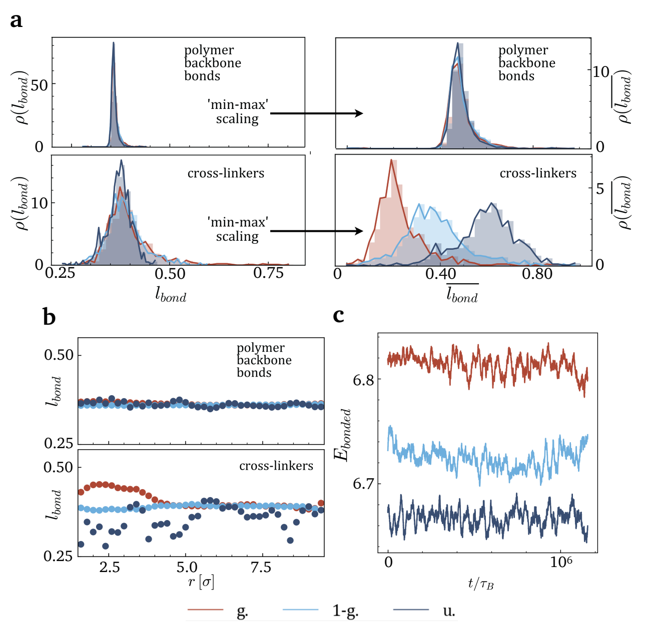

Fig. 1 a (left) Histograms of l_{bond} distributions for polymer backbone stretchable springs (FENE potentials) and cross-linkers (harmonic potentials). (right) Histograms of 'min-max' normalized bond length $\overline{l_{bond}}$ for various morphologies. b Average l_{bond} as a function of the distance r from the center of mass of the MNG. c Energy of bonded interactions within various morphologies. Along the x-axis is the simulation time divided by the Brownian relaxation time of a MNP. Both l_{bond} and r are measured in units of bead diameter, σ.

2 Fitting Magnetisation Autocorrelation Functions (ACF)

As mentioned in the main text, the shape of the MNG ACFs is rather complex and required 10 stretched exponential functions $e^{-t^{\beta}}$ for exponents $0 < \beta < 2$ to fit them in a reliable way. We employed Differential Evolution algorithm in order to optimise the fitting. Obtained weights of the stretched exponentials are provided in the following three tables.

Table 1 gaussian

i	f_i	τ_i	β_i
0	0.0313833796	668869.846	1.94976674
1	0.0571745925	27.3872559	0.553795715
2	0.0272604858	573313.803	0.218793286
3	0.122433377	673301.266	1.94916583
4	0.334049327	311.535839	0.451347345
5	0.0463671472	664291.87	1.94998642
6	0.113471535	11.6346341	0.776222096
7	0.0777341818	120532.938	1.94999992
8	0.0425189368	44376.0054	1.04497825
9	0.150850617	5.54432802	0.542271495

Table 2 1-gaussian

i	f_i	τ_i	β_i
0	0.134381915	345126.302	1.94989022
1	0.052857027	1550.03396	0.747324177
2	0.132876636	7.2360366	0.594977791
3	0.116992815	29.1948474	1.63578785
4	0.0831295954	344203.601	1.94972976
5	0.0862043751	2.94987194	0.482914566
6	0.0580794413	38670.5705	1.38074828
7	0.103970802	1522.14379	0.62283344
8	0.0716531728	345811.168	1.94999376
9	0.162747132	158.250851	0.556393399

Table 3 uniform

i	f_i	τ_i	β_i
0	0.0348141118	503704.753	1.95
1	0.0735062133	468.654362	1.75026195
2	0.00787281569	63653.0372	1.52948371
3	0.114478161	120.786526	0.280530386
4	0.1288664	89.9251735	1.53562632
5	0.0841476288	33.1085893	1.94994946
6	0.0808016172	3864.01534	0.422685835
7	0.223120481	503093.708	1.94998276
8	0.021686283	56871.6425	1.40049073
9	0.234999511	3.49137971	0.806357891

3 Tracer Diffusion

In order to obtain the escape time and the diffusion coefficients of the tracers we calculated the mean-squared displacement for the three considered morphologies, and field scenarios. The results are presented in the three figures below as indicated in the corresponding legends.

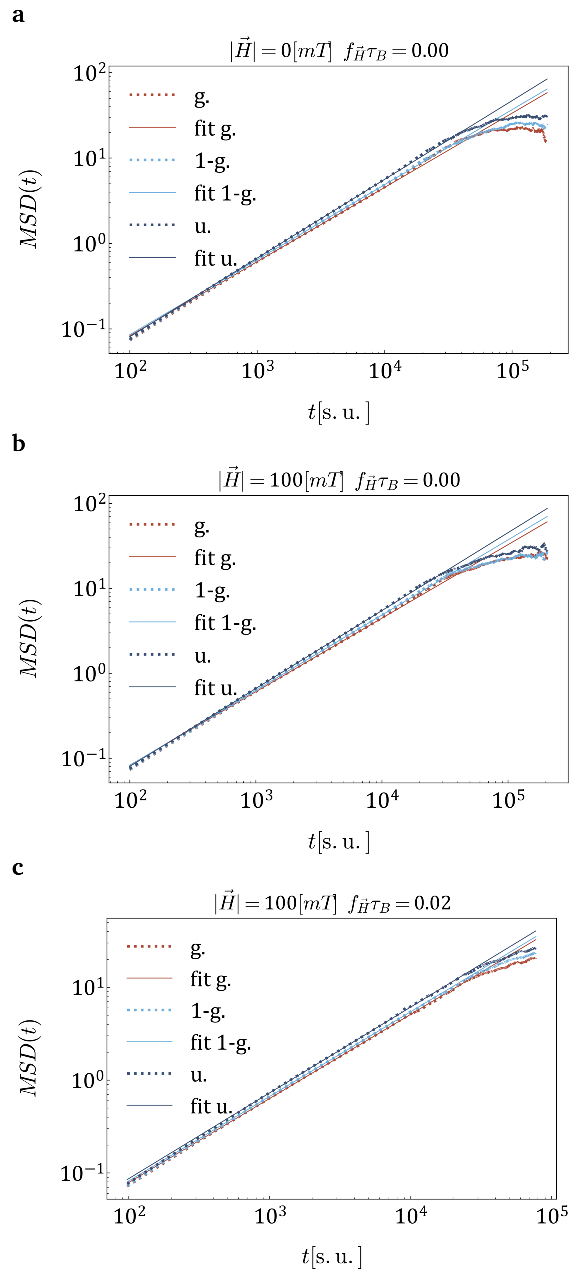

Fig. 2 $MSD(t)$ a Zero-field case. b Constant non-zero-field scenario. c Rotational-field situation.