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Particle-Matrix Coupling in Magnetic Gels: Simulations and Experiments

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The stimuli-responsive behavior of magnetic gels depends critically on the coupling between magnetic nanoparticles (MNPs) and the surrounding polymer matrix. However, the influence of competing effects such as hydrodynamics, van der Waals forces, electrostatics, and local network structure on the gels' behavior are not fully understood: in many cases, the nanoparticles are not covalently attached to the polymer network but are instead physically trapped within its meshes. Despite this weak mechanical coupling, experiments reveal signatures of hindered rotational dynamics.

We address this by means of AC susceptometry, which is available experimentally and in simulations. The technique probes the MNPs response to AC magnetic fields and provides information about the coupling between MNP rotation and the MNPs' local environment. In our study, we juxtapose measurements on PAAm hydrogels loaded with cobalt ferrite MNPs with coarse-grained simulations that explicitly resolve particles, polymers, and hydrodynamic interactions. Experimentally, the mesh size is tuned via the degree of crosslinking and the monomer fraction; in simulations, in addition to the mesh size, the MNP–polymer interaction is varied considering both, isotropic and anisotropic couplings.

In simulations, we observe that hydrodynamic interactions between the MNP and the surrounding polymer shift the susceptibility spectrum towards lower frequencies. The magnitude of the shift depends on the density of polymers close to the MNP, which is higher, for smaller polymer mesh sizes but also if there is an attraction between the MNPs and the polymer. Hence, a strong isotropic MNP–polymer affinity can mask mesh-size effects, by distorting the surrounding polymer network.

Experimental spectra show a systematic decline of the low-frequency susceptibility with decreasing mesh size, even though the MNPs are only physically trapped, not covalently crosslinked. Purely hydrodynamic coupling or isotropic Lennard-Jones-type attractions fail to reproduce this drop in simulations. Reproducing the experimentally observed susceptibility reduction requires introducing anisotropic, patchy attractions of a few kT between the MNP surface and the polymer chains. Thereby, our results indicate that alongside hydrodynamic and isotropic van-der-Waals like interactions, surface heterogeneity of the MNPs can be a relevant factor governing MNP–matrix coupling, even if the MNPs are not covalently bound to the polymers.

Type:

Oral