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Tuning the Viscoelastic Properties of Hydrogels via Surface-Engineered Iron Nanoparticles

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The macroscopic properties of magnetic soft matter are fundamentally governed by the microscopic interactions at the nanoparticle-matrix interface. While magnetite is the standard in magnetic fluids, nanoscale zero-valent iron (nZVI) offers a superior magnetic profile, with a saturation magnetization of approx. 220 emu/g, that significantly enhances the magnetic torque and responsiveness of the composite. However, the high surface energy and chemical instability of nZVI require sophisticated surface engineering to prevent aggregation and control its integration into self-assembled networks.
In this work, we investigate the physico-chemical characterization of nZVI nanoparticles functionalized with hydroxyl- and amine-terminated PEG chains (NH_2-PEG and APTES). XPS and zeta potential measurements confirm the formation of a dense, stable polymer shell that modulates the surface charge and chemical potential of the nanoparticles. These engineered MNPs were embedded into a supramolecular hydrogel matrix formed by the self-assembly of fluorenylmethoxycarbonyl-diphenylalanine (Fmoc-FF) peptide sequences.
We demonstrate that the surface chemistry of the MNPs acts as a structural template during the pH-triggered self-assembly process. Rheological characterization reveals that even at an ultra-low concentration of 0.3 vol%, the functionalized nZVI particles significantly alter the storage modulus of the network, indicating a strong coupling between the nanoparticle surface moieties and the peptide nanofibers. Furthermore, we evaluate the biological interface through hemostatic assays, showing that the surface-induced interactions can either promote or inhibit blood coagulation. This study provides fundamental insights into how fine-tuning the nanoparticle interface allows for the rational design of injectable, high-Ms magnetic nanocomposites for advanced biomedical and technological applications.
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