Supplementary Information (SI) for Soft Matter.

Supplementary Material

Magnetorheological study of uncoated nanoparticles dispersed magnetic ionic liquid

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1. Atomic force microscopy (AFM) measurment

To analyze the particle size distribution, AFM images are shown in Figure S1 (A) and Figure S1 (B) which are obtained from small-sized magnetic nanoparticles (S-MNPs) and large-sized magnetic nanoparticles (L-MNPs), respectively.

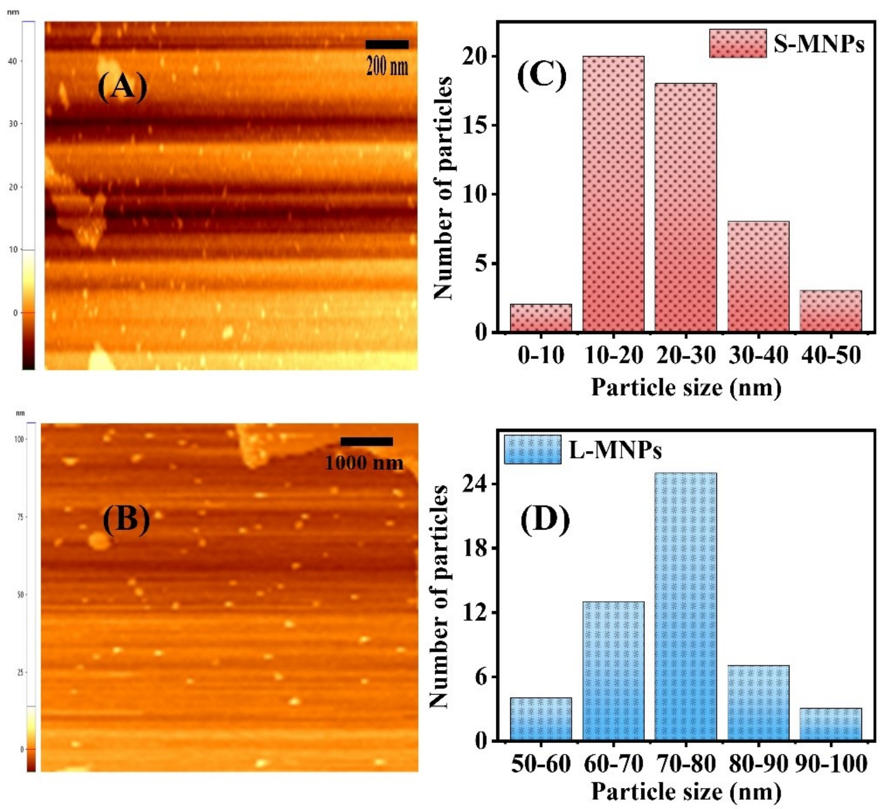


Figure S1: (A) Atomic force microscopy (AFM) images of small-size magnetic nanoparticles (S-MNPs) and (B) large-size magnetic nanoparticles (L-MNPs) (C) particle-size distribution histogram for S-MNPs and (D) particle-size distribution histogram for L-MNPs.

A sparse quantity of nanoparticles (10^{-6} wt%) was dispersed in water to prepare a highly dilute solution. The solution was then drop-cast onto a clean silicon substrate to let them dry before AFM imaging. The AFM imaging was performed in non-contact mode. The histograms shown in Figure S1 (C) and Figure S1 (D) provide the distribution of particle size for S-MNPs and L-MNP respectively. While the S-MNPs have a size less than 50 nm, the L-MNPs are above 50 nm.

2. Linear viscoelastic region (LVR)

LVR is the range in which reproducible oscillatory measurements can be made without causing structural damage to the sample. It measures the storage (G') and loss (G'') moduli as a function of applied strain (γ). This is known as amplitude sweep measurement. Figure S2 shows these measurements for various samples at different compositions. Further, the data have been collected in the absence (B: 0T) and presence (B: 1T) of magnetic field. A shear strain of 0.1% appears to be the common shear strain for all the samples which is within the LVR.

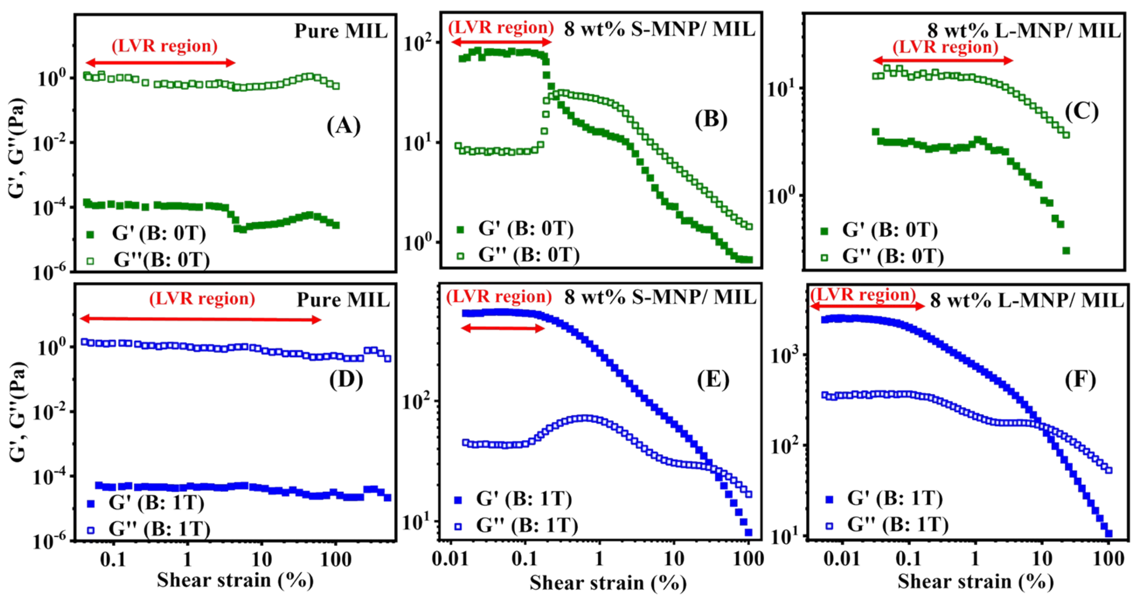


Figure S2: Variation of storage modulus (G') and loss modulus (G'') of (A) pure magnetic ionic liquid (MIL), (B) MIL with 8 wt% of small-size magnetic nanoparticles (S-MNPs) and (C) MIL with 8 wt% of large-size magnetic nanoparticles (L-MNPs) without any applied magnetic field (B = 0 T). (D), (E) and (F) show the respective data in the presence of 1 T magnetic field.

3. Non-Newtonian behaviour of magnetic ionic liquid (MIL) with added magnetic nanoparticle (MNP)

As shown in Figure S3, the viscosity of pure MIL is constant over the whole range of shear rate indicating it to be a Newtonian fluid. However, in presence of MNPs, the fluid becomes non-Newtonian showing shear thinning behaviour. An interesting observation is that at a given shear rate, the viscosity of MIL with smaller MNP (S-MNP) is higher than the larger MNP (L-MNP). At the same wt.%, the number density of S-MNP is higher than the L-MNP. Therefore, there will be more particle-particle and particle-liquid interactions which may cause higher friction during fluid flow. At low shear rates, MNPs are more likely to form local microstructures or aggregates leading to a pronounced increase in viscosity. As the shear rate increases, these microstructures begin to align, resulting in shear-thinning behaviour.

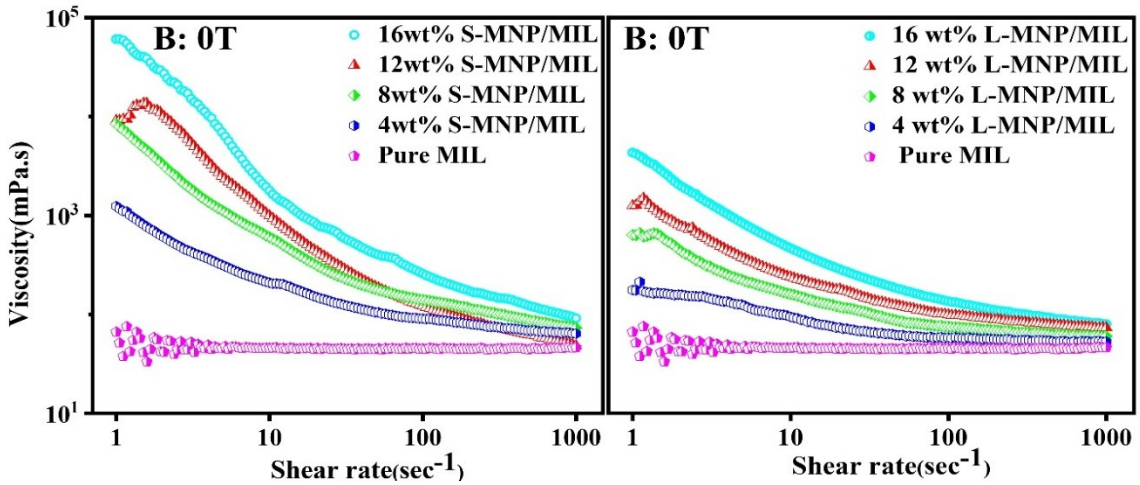
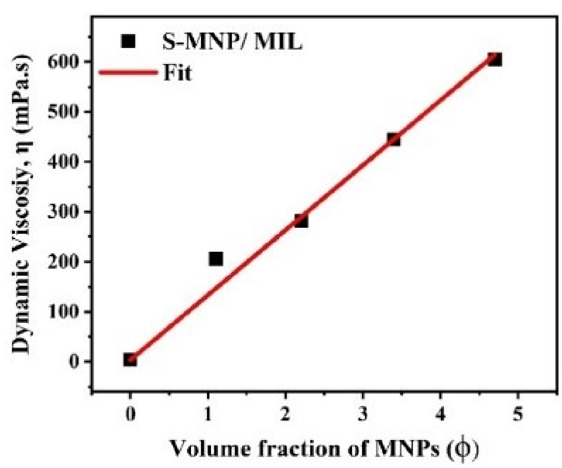


Figure S3*: Viscosity versus shear rate curves for pure magnetic ionic liquid (MIL) in the presence of different concentrations of (A) small-size magnetic nanoparticles (S-MNP) and (B) large-size magnetic nanoparticles (L-MNP) without a magnetic field.*

4. Viscosity of MIL-MNP solution with concentration of MNP

For a given magnetic field of 0.2 T, the viscosity (η) of the small-size magnetic nanoparticle (S-MNP) dispersed magnetic ionic liquid (MIL) is shown in figure S4. As evident, η increases linearly with the nanoparticle volume fraction (ϕ). The data have been fitted with Einstein viscosity expression, $\eta = \eta_0(1 + \alpha_f\,\phi)$, where η_0 is the viscosity of pure MIL which has a of 3.6 mPa.s. Here, α_f is the fitting parameter with a value of 35. This value is quite high compared to classical systems where there is no strong interaction between the solvent and the dispersed particle.



FigureS4: Viscosity of dispersion of magnetic nanoparticle in magnetic ionic liquid with the volume fraction of the particle. The data (filled squares) have been fitted with Einstein viscosity equation (red solid line).