Supplementary material

S1. Redispersibility tests

This work aimed to demonstrate that the polydispersity of carbonyl iron particles described by particle size distributions with the same mode (~ 7 μm), but different distribution widths, affects the maximum packing of these particles.

During the preparation of the MRF, three powders were mixed, always in the same volume fraction of solids (48.5 vol%), and their difference was only the width of the histograms. As expected, the off-state shear viscosity is reduced with increasing polydispersity due to an increase in the maximum packing. However, contrary to what is often reported in the literature, an increase in polydispersity also increased the yield stress under a magnetic flux density B = 0.57 T, measured at the controlled shear stress ramp test.

The redispersibility of each sample was measured after 30 days at rest with natural sedimentation and under normal gravity. In addition, each sample was allowed to sediment under different periods (For more details on this test, we recommend the reference Gomes de Sousa[13].

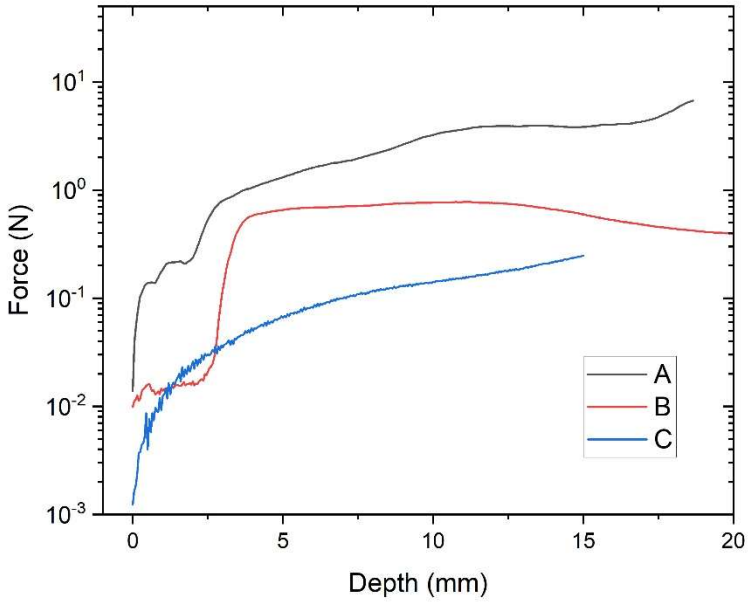

Figure S1 - Normal force of each MRF sample, after one month of settling under normal gravity, as function of the penetration of the blade penetration. All tests were executed at room temperature.

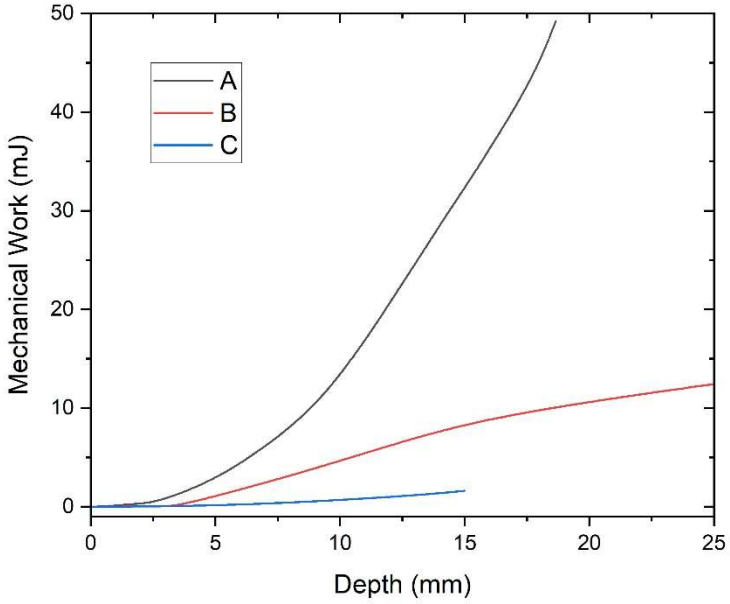

Figure S2 - Mechanical work spent by the rheometer, as a function of depth, for each MRF sample. The work was calculated by simply integrating the area under the Force x Depth curves.

From Figure S1, one can see that sample A had the worst performance against redispersibility: the normal force reached 6.7 N at a depth of approximately 19 mm. Sample B, in turn, had an intermediate performance, with a maximum force of 0.8 N at a depth of about 10 mm. Finally, the best performance in this test was that of sample C, whose maximum normal force was 0.25 N at 15 mm depth.

Figure S2, in turn, shows that the mechanical work spent during the blade penetration in each sediment was about 1.6 mJ, 8.3 mJ, and 32.4 mJ at the same 15 mm depth for samples A, B, and C, respectively.

These results confirm that an increase in polydispersity was advantageous not only for the off-state viscosity and on-field yield stress but also for MRF formulations with better performances against redispersibility.

S2. Estimating the magnetic flux density (B)

During the rheometry measurements, the MRD cell used to obtain the yield stress values under an applied magnetic field was the first commercial version from Anton Paar - Physica GmbH (Germany). In this first version (2001 – 2002), the external magnetic field strength H is measured by placing a Hall probe inside the gap (see figure S3). In this configuration, with a direct current (DC) of I = 2 Amperes applied to the coil of the MRD cell, an external magnetic field of H = 270 kA/m is measured.

Figure S3 - MRD-180 cell (Anton Paar, 1st version, 2002) with the Hall effect probe inside the gap between the plates, power supply DC (I = 2.00 A), and Teslameter "Magnet-Physik" FH-54 showing **H** = 0.270 MA/m (or 270 kA/m). The gap was empty, in air, and with no MRF sample inside because it is not possible to directly measure the magnetic flux density **B** with the gap filled with MRF samples, in this MRD cell configuration.

The magnetic flux density B in the MRF sample can be estimated by applying the Wollny *et al.* (2002) [84] equation:

$$B = 7.539 \times 10^{-4} \cdot K \cdot \frac{I}{0.0035 - \frac{d}{[m]}\left(\frac{\mu_r - 1}{\mu_r}\right)} \cdot \left[\frac{T}{A}\right] \quad (1)$$

In this equation, according to the authors: "*As the relative permeability μ_r of the sample MRF is not a constant but depends on the magnetic field strength, the calculation has to be done iteratively. Magnetic Flux Density in [T], I is the current in Ampere [A] and d denotes the gap size in meter [m]. The Calibration Factor K is an adjustment parameter dependent on the geometry. Here, this parameter equals one for the measuring system being used (20 mm diameter parallel-plate and 1 mm gap)*." [84].

However, to use such an equation, it is necessary to know:

1. The value of the electric current applied to the coil of the MRD cell (which is easy to measure).
2. The constant K, which is equal to one if the plate used has a diameter of 20.0 mm and the gap = 1 mm (exactly the dimensions we use).
3. The relative magnetic permeability μ_r of the MRF sample that was inserted into the gap.

This last parameter, μ_r is more complicated to compute. Only with a magnetometer or some similar device it is possible to measure this value exactly.

Fortunately, it was possible to estimate μ_r for our MRF mixtures from the values reported by BASF for two of their commercial MRFs: Basonetic 2040 and 5030, which have, respectively, ϕ = 20 and 50 vol% of carbonyl iron powder, and whose relative permeabilities are μ_r = 3.98 (20 vol%) and 7.96 (50 vol%). As for pure polyalphaolefin oil, without any ferromagnetic material (ϕ = 0 vol%), we have μ_r = 1. Assuming a linear behavior of μ_r with an increase in the volume fraction of solids (carbonyl iron powder), it was possible to construct the graph in Figure S4 and find the linear regression equation of $\mu_r = f(\phi)$ with a correlation coefficient of adjustment R^2 = 0.99793.

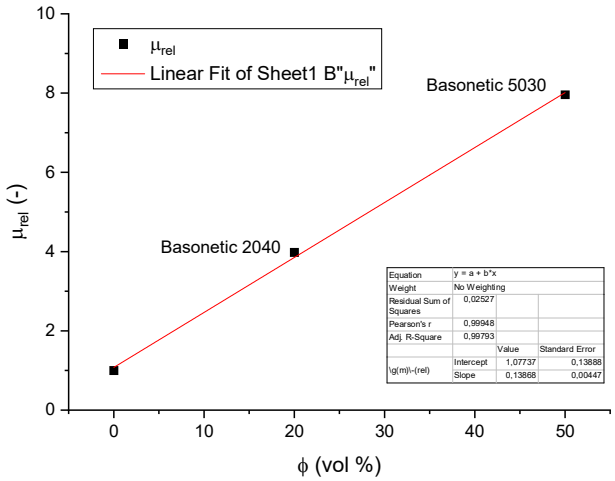

Figure S4 – The relative permeability as a function of the volume fractions of iron powder for pure PAO oil (0 vol%), Basonetic 2040 (20 vol%) and Basonetic 5030 (50 vol%).

The resulting linear equation (with ϕ in vol%) is: $$\boldsymbol{\mu_{rel} = 0.13868 \cdot \phi + 1.07737} \quad (2)$$

Since all 3 mixtures have the same volume fraction of ϕ = 48.5 vol%, we estimate a value of μ_r = 7.80 for our MRF samples.

With this value of μ_r and applying equation (1) from Wollny *et al.* (2002) [84] with I = 2.0 A and K =1, we can finally estimate a magnetic flux density value **B = 0.57 Tesla**, with the MRF of the 3 mixtures.

[84] Wollny K, Läuger J and Huck S. Magneto Sweep – A New Method for Characterizing the Viscoelastic Properties of Magneto-Rheological Fluids. Appl. Rheol. 2002; 12(1): 25-31. DOI: 10.1515/arh-2002-0003.