## **Electronic supplementary information**

# High performance magnetorheological fluids: very high magnetization FeCo-Fe<sub>3</sub>O<sub>4</sub> nanoclusters in ferrofluid carrier

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#### Fe<sub>3</sub>O<sub>4</sub>/OA- FeCo/Al<sub>2</sub>O<sub>3</sub> MNCs

The magnetic clusters (6) based on combination of OA surfacted  $Fe_3O_4$  nanoparticles (1) with FeCo /  $Al_2O_3$  particles (2), coated with a hydrophilic layer of sodium lauryl sulfate (SLS) surfactant (3) were prepared as follows.

Pipette 915  $\mu$ l of hydrophobic magnetic fluid into a Berzelius beaker, corresponding to 0,5 g of OA coated magnetite nanoparticles Fe<sub>3</sub>O<sub>4</sub>/OA (1) dispersed in toluene as carrying organic solvent (4). To this solution different amounts of FeCo / Al<sub>2</sub>O<sub>3</sub> particles (2) are added, corresponding to a mass ratio (wt%) in the range 25 – 85 %. The addition of FeCo / Al<sub>2</sub>O<sub>3</sub> particles is done in 4 - 5 doses simultaneously with the addition of 0.3 ml of toluene for emulsifying. The particles mixture was mechanically mixed using a glass rod until the FeCo / Al<sub>2</sub>O<sub>3</sub> particles (2) are completely incorporated and the remaining toluene was added to a final toluene volume of 2 ml.

Separately in a Berzelius beaker dissolve the required amount of SLS (3) (1.795 g, 1.75%), in 100 ml of distilled water. In that samples where higher concentrations of FeCo /  $Al_2O_3$  particles are added, the concentration of SLS is increased proportionally with the amount of FeCo /  $Al_2O_3$  particles added, in order to create and stabilize the mycelium formed in the miniemulsification process. In the aqueous solution of SLS surfactant (3), the organic solution of Fe<sub>3</sub>O<sub>4</sub>/OA nanoparticles (1) and FeCo /  $Al_2O_3$  (2) particles mixture was added under mechanical stirring. The biphasic mixture, oil in water, is ultrasound using an ultrasonic finger in order to obtain the aqueous phase organic emulsion (5). The ultrasound is performed for 2 minutes, at an amplitude of 50%, at room temperature.

The as formed emulsion (5) is transferred to a higher Berzelius beaker and kept in an oil bath under strong magnetic stirring (600 rpm) for 30 minutes, at a temperature of 90 °C, to evaporate the organic solvent, toluene (3), which is volatile at this temperature. At the end of the solvent evaporation time, the sample is magnetically separated from the reaction medium by placing the beaker on a neodymium cubic magnet, and the reaction medium containing residues of reactants or by-products is discharged.

Finally, the sample is washed three times with a water-methanol mixture to remove traces of toluene. Finally, the magnetic cluster (6) obtained in a fine powder form is dried for further measurements or redispersed to a known concentration in aqueous medium.

#### Magnetometry



**Figure S1**. Hysteresis loops of dried a)  $Fe_3O_4$  MNPs, b)  $FeCo/Al_2O_3$  MNPs, and c)  $Fe_3O_4$ - $FeCo/Al_2O_3$  MNCs, at 10K, 100K, 200K and 300K. A central field interval is considered in the inset for a better evidence of the variation of the coercive fields.

#### Viscosity curves



### (b) MRFF50-40% and FF500

**Figure S2**. Viscosity curves for different values of the magnetic induction B (empty symbols-FF, full symbols-MRFF): (a). MRFF500-10% and FF500; (b) MRFF500-40% and FF500.

#### Magnetoviscous (MV) effect



(b)

**Figure S3.** Dependence of magnetoviscous effect on the magnetic induction B, for two values of the shear rate: (a)  $\beta = 8.73 s^{-1}$ ; (b).  $\beta = 1000 s^{-1}$ .