Formation of particle clusters from rotating particle chains

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Superparamagnetic particles do only show a magnetization, as long as an external magnetic field is applied. In a homogenous magnetic field, the particles only interact with each other. This interaction leads to chains of particles (see Figures 3a,b) [1]. If the field begins to rotate slowly, the chains will begin to rotate. Dissolved in a fluid, like water, the chains break apart when higher rotational frequencies are used. Depending on particle concentration, rotation frequency and the applied field’s strength, the particles will form highly ordered structures (see Figures 3d,4) [2]. There are a few theoretical models for the formation and breaking of particle chains like [3], but the formation of particle clusters at high rotation frequencies is, to our knowledge, not been modelled before. Therefore, a COMSOL Multiphysics® model is presented to study their formation and hopefully improve the processes involved.

The demagnetization fields of the particles are assumed to be similar to that of a homogenously magnetized ball. As this is inhomogeneous, other particles are subject to a force of \( \mathbf{F} = \mathbf{\mu}_0 \mathbf{M} \cdot \nabla \mathbf{B}_\text{ext} \).

The magnetic moments \( \mathbf{m} \) of the particles are approximated to be aligned with the external field \( \mathbf{B}_\text{ext} \) and equal to \( \mathbf{m} = V \mathbf{m}_0 \mathbf{B}_\text{ext} \) with the particle volume \( V \) and the particle’s magnetic susceptibility \( \chi \). Taking everything into account the magnetic force is [3]

\[
\mathbf{F}_m = \frac{3}{2} \mu_0 \mathbf{m} \cdot \nabla (1 - 5(\mathbf{m} \cdot \mathbf{r})^2 + 2(\mathbf{m} \cdot \mathbf{r})^2) \mathbf{m}
\]

Figure 1 Plot of the force in the center exerts on an entire particle, when both have a vertical magnetization. Blue marks a high drag, yellow a high push. The black line marks the point where no force towards the particle is exerted.

The counteracting force is the Stokes drag, which will cause the chains to break apart \( \mathbf{F}_S = 6 \nu V \text{viscosity} \mathbf{r} / (\nabla \mathbf{v}_\text{fluid} - \nabla \mathbf{v}_\text{particle}) \).

All other effects, like Brownian motion and the particles’ influence on the fluid, are omitted to keep the model simple.

The COMSOL Multiphysics® interface “Particle Tracing for Fluid Flow” is used to handle the simulation. The “Drag Force” node, set to “Stokes”, accounts for the Stokes Drag. The magnetic force is implemented as an “particle-particle interaction” node with an user-defined force. To prevent the particles from overlapping another “particle-particle interaction” node with a “Lennard-Jones” option is used, where the attracting part of the force is switched off manually. The standard solver settings are used and the time steps are set to 1ms. The computation is quite time consuming.

The particles are placed in a homogeneous, rotating magnetic field created by a rig of four Helmholtz coils (see Figure 2). The particles are observed with an optical microscope.

An in-depth discussion of the experimental methods and results may obtained in [4]. All Experiments where carried out with superparamagnetic particles with a size of upto a couple of micrometers.

The first simulation was carried out with more or less randomly chosen parameters, to check for the behavior observed in the experiments.

Figure 2 Rig of Helmholtz coils used to produce a homogeneous, rotating magnetic field [4]

Figure 5a Two particle chains slowly rotating. The magnetic field’s orientation is indicated by the red arrows

Figure 5b At higher rotation frequencies the chains deform into s-shape.

Figure 5c The particles break apart into smaller chains, which are again able to follow the field’s rotation

Figure 5d The particles form a highly ordered cluster which is slowly rotating around its center of mass.

The Figures 5a-d show that a behavior similar to the experiment can be achieved with the model used.

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