Magneto-Granulometric Analysis of Concentrated Ferrofluids

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The determination of particle size distribution in magnetic fluids is normally performed either using an electron microscope [1] or analyzing the magnetization curves (magneto-granulometric analysis) [2-4]. The first method allows direct measurements but requires treating a lot of particles (10^3 – 10^4). The latter is more suitable and efficient as it gives information on both particle dimensions and magnetic properties of ferrofluids. That is why the analysis of magnetization curves is frequently employed to estimate the sizes of single-domain particles.

This study is focused on the adequacy of various theoretical models in determination of particle size distribution during processing of magnetization curves for concentrated ferrofluids. It was first demonstrated in Ref. [3] that the choice of theoretical model significantly influences the results of magneto-granulometric analysis. In this paper the following idea was suggested. The best theoretical model must be chosen on the basis of experiments on solutions differing in concentration but having the same granulometric composition. The influence of interparticle interactions on magnetization increases with particle concentration. Therefore, the independence of magneto-granulometric analysis results for concentration is a necessary condition of the model adequacy in describing interparticle interaction and magnetization curves.

Solutions of different concentrations were obtained [3] by diluting base ferrofluid with kerosene. Identical granulometric composition was ensured automatically provided that solutions did not segregate and particles did not presipitate from solution. The base „magnetite in kerosene“ ferrocolloid of saturation magnetization 57 *kA*/*m* was obtained under laboratory conditions using the chemical condensation technique and had high sedimentation stability. Six additional samples were prepared differing in degree of dilution $n = V_1 / (V_1+V_2)$ of the base ferrocolloid. Here, V_1 and V_2 are the volumes of the base ferrocolloid and kerosene, respectively.

The results of magneto-granulometric analysis are presented in Tables 1 and 2. The particle distribution over the magnetic core diameter is described by the gamma-distribution [3,4], and the following models are studied: the one-particle Langevin model, the mean field model by Weiss, the mean spherical model (MSM, [5]), the high temperature perturbation model (HTA, [6]); the modified mean field model of the 1st (MMFM-1, [3]) and 2nd (MMFM-2, [4]) orders, Born-Mayer cluster expansion theory (CET, [7]) that provides exact results up to a certain order of the interaction parameter λ and the volume fraction ϕ.

When using the one-particle Langevin theory, the calculated value of the particle size distribution width increases with the growth of particle concentration. This increase is a natural result of interparticle interactions. Extension of the particle size distribution has the same impact on the magnetization curve as the dipole-dipole interaction, thus masking the effect. Moreover, for most diluted sample all models, taking into account the interparticle interactions, give the same results, except the one-particle Langevin theory. (That the mean diameters resulting from CET are consistently smaller is caused by using in the CET a discretized gamma distribution: doing the same, e.g., in HTA reduces the values accordingly.)

The reason is clear: the dipole-dipole interaction results in the appearance of demagnetization fields. Their influence may be very weak for diluted ferrofluids, but they exist. It means that the Langevin theory is absolutely inapplicable even to diluted ferrofluids.

Model	Degree of dilution (*n*)						
	0.088	0.137	0.197	0.296	0.444	0.664	1
Langevin	0.44	0.45	0.47	0.54	0.57	0.60	0.64
Weiss	0.41	0.40	0.39	0.40	0.36	0.31	0.25
MSM	0.41	0.41	0.41	0.44	0.44	0.44	0.45
HTA	0.41	0.41	0.42	0.46	0.47	0.48	0.55
MMFM-1	0.41	0.41	0.42	0.45	0.45	0.44	0.46
CET	0.418	0.420	0.418	0.417	0.418	0.412	0.382
MMFM-2	0.410	0.410	0.409	0.409	0.410	0.410	0.410

Tab. 1: Width of particle size distribution.

Model	Degree of dilution (*n*)						
	0.088	0.137	0.197	0.296	0.444	0.664	1
Langevin	7.1	7.2	7.0	6.4	6.3	6.3	6.0
Weiss	7.3	7.6	7.7	7.6	8.0	8.5	8.9
MSM	7.3	7.5	7.5	7.1	7.2	7.2	6.9
HTA	7.3	7.4	7.4	6.9	6.7	6.5	5.6
MMFM-1	7.3	7.5	7.5	7.1	7.1	7.1	6.8
CET	7.13	7.13	7.13	7.19	7.24	7.46	8.25
MMFM-2	7.30	7.32	7.32	7.31	7.30	7.30	7.29

Tab. 2.: Mean particle diameter (*nm*).

The Weiss model predicts a higher interparticle interaction intensity than in real ferrofluids. The HTA describes the initial and final segments of the magnetization curves fairly well but overestimates the magnetization in the middle segment. That is why, the HTA results prove to be just slightly better than those obtained using the Langevin model. More stable results are obtained with the help of MSM and MMFM-1. In these cases the monotonic variations of the mean particle diameter and the distribution width do not exceed 10 %. The CET [7] gives even better results but fails for the highest concentration where higher orders of λ and ϕ than those taken into account give nonnegligible contributions. The deviations in the predictions of MMFM-2 [4] being less than 1 % are the smallest.

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