SELF-ASSEMBLED PARTICLE LAYERING INDUCED BY ELECTRIC FIELD IN TRANSFORMER OIL-BASED FERROFLUID BY NEUTRON REFLECTOMETRY

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Understanding the physical-chemical processes during magnetic nanoparticles (MNPs) assembly and manipulation of the assembly process is highly important for synthesis of well-defined architectures from a nanometer scale and thus fabrication of such novel functional nanostructures will open horizon for new materials and devices. Assembly could lead to controllable NPs arrangement in bulk or at interfaces and it can be achieved just by using some template or external stimuli such as magnetic or electric fields. At interfaces, a uniform magnetic field amplifies the dipole-dipole interaction between MNPs in ferrofluids (due to one-direction alignment of particle magnetic moments) which enhances the layering of MNPs on a planar surface. The regulating properties by external electric fields for dielectric ferrofluids was recently reported based on the small-angle neutron scattering studies of a transformer oil-based ferrofluid (TOFF) showing bulk structuring and phase separation under electric fields [1]. The nanofluids are of practical importance - especially they have been used in several thermal management systems, as they contribute to the augmentation of the heat dissipation rates in many applications. The theoretical consideration [2] shows that sufficiently strong applied electric fields cause non-uniform distributions of particles between electrodes. The macroscopic structural changes in a simple ferrofluid consisting of iron oxide nanoparticles coated with oleic acid and dispersed in transformer oil under electric field have been proved [3]. Effects of magnetic field on ferrofluids at planar interface were revealed by neutron reflectometry (NR) based on the analysis of the evolution of specular reflectivity [4, 5].

The aim of this work is to investigate the interface structural changes in TOFF with magnetite nanoparticles as well as the magnetic fluid-solid interfaces under electric fields. An important question is whether electric fields, similar to magnetic fields, could be a driven force to induce the assembly of magnetic nanoparticles at the interface and whether formation of additional layers in ferrofluids at the inner surface of transformer could increase dielectric breakdown voltage. Specular reflectivity of non-polarized neutrons was measured at the neutron reflectometer GRAINS with a horizontal sample plane configuration installed at the IBR-2 pulsed reactor of JINR (Dubna, Russia). Assembling of superparamagnetic NPs of a dilute classical ferrofluid - magnetite coated with oleic acid in transformer oil – on a planar surface of the metal electrode (copper) was observed when an out-of-plane electric field is applied to the interface. The obtained NR curves were processed in a standard way in terms of the Parratt formalism using the Motofit package for the IGOR Pro software.

For the initial state (no electric field) the best fits are obtained when assuming one ‘wetting’ layer of MNPs on the surface. The corresponding SLD profile is presented in Fig. 1a. The reflectivity curves for the interface under electric field were fitted with all fixed values of the solid components parameters and all vary parameters of FF components. The electric field induced evolution of the ‘liquid part’ of the interface in terms of the changes in the SLD profile is followed in Fig. 5b. As expected, the increase in the electric field slightly affects the structure of 1-st layer determined by the surfactant shell; one can see small reduction in thickness, roughness and SLD of this layer. More significant changes are observed for the 2-nd layer. It becomes thicker and more concentrated thus evidencing the enhanced adsorption of MNPs. The layer thickness growths with the electric field increase together with the SLD. Starting from 300 kV/m, the fits of the reflectivity curves are better if an additional layer #3 is introduced. At maximum field (700 kV/m) it becomes even higher. On the one hand, there is a tendency towards a saturation (regarding the content of MNP) of the two adsorption layers with the electric field increase. At the same time, some redistribution of MNP in the two layers takes place at maximum field intensity, reflecting a decrease of the MNPs content in the first layer. NR curves were also measured in different moments during 9 hours after the field of 700 kV/m was reduced to zero. It is interesting that the adsorbed layers evolve to some extent after the external field is switched off.

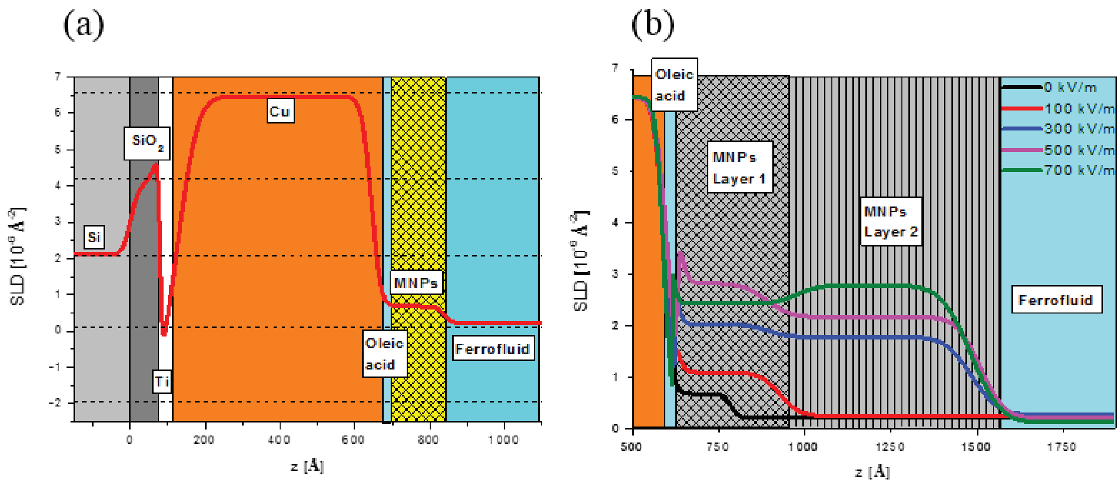

Figure 1. SLD depth profiles (plotted as a function of the distance z from the substrate surface) determined from the fits to the NR data (a) SLD profile in full z-range for zero field (initial configuration). For comparison, theoretical SLD values for the interface components are shown by dashed lines. (b) SLD profiles at the electrode-ferrofluid interface at different intensities of the external electric field.

The reason of the observed effects is related to the polarization of the particles in the electric field and their inter-action as dipoles. The observed self-assembled layering could be used as an additional barrier at the inner surface of transformer to increase dielectric breakdown voltage of working fluids.

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