Ultrasonics 55 (2015) 26–32

Contents lists available at ScienceDirect

Ultrasonics

journal homepage: www.elsevier.com/locate/ultras

Effect of carrier and particle concentration on ultrasound properties of magnetic nanofluids

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ARTICLE INFO

Article history:
Received 3 June 2014
Received in revised form 7 August 2014
Accepted 12 August 2014
Available online 23 August 2014

Keywords:
Magnetic fluid
Ultrasonic wave propagation
Nanofluid

ABSTRACT

Ultrasound wave propagation in nanofluids and its rheological behavior has been studied as a function of solid volume fraction, temperature and magnetic field for magnetic nanofluids synthesized in kerosene and transformer oil. Ultrasonic velocity decreases while viscosity increases with increasing volume fraction. The attenuation of ultrasonic wave is explained using dipolar coupling co-efficient which favors oligomer structures with increasing number density of particles. The structure formation increases further with increase in magnetic field which is prominent in transformer oil compared to kerosene. This difference can be due to the structural difference between these two carriers.

1. Introduction

Magnetic nanofluids represent a technologically important material because of its wide scope of tuning the macroscopic behavior under different environment. Recent experiments and analysis show that magnetic dipole force and strong magnetic field expels nanoparticles to form chains and aggregates that can greatly affect the macroscopic properties of ferrofluids even for low concentration [1–6]. The formation of structural pattern in the magnetic fluid and its thin film under the influence of magnetic field can be investigated by optical microscopy, strong light diffraction image, electron microscopy, nuclear magnetic resonance (NMR) technique, small-angle neutron scattering (SANS), acoustical study and rheological studies [7–16]. This clustering has a significant influence on the properties of magnetic fluid and hence its further use.

In 1975, Hayes had reported needle-like clusters or aggregates of the magnetic particles in nanofluids through optical microscopic observations under the influence of an external magnetic field [7]. Though magnetic nanofluids is an opaque liquid when it is sandwiched by two glass slides and pressed to a thin film of a few tens of micron thickness, visible light can be transmitted through the magnetic nanofluids film. The alternate study is to project the transmitted light on screen and obtain a peculiar scattering light pattern. Haas and Adams [8] observed a peculiar projection of the transmitted light from the diluted magnetic nanofluid's film when the field was perpendicular to the light propagation. The needle-like clusters in the magnetic nanofluids play a role of grating of an optical diffraction grating. The strong diffraction effect is due to the periodical spacing between the needlelike clusters. On the contrary, by measuring the diffraction light intensity as a function of the diffraction angle, one can calculate the spacing of the clusters by Fourier transform [9].

The lower limit of an ordinary optical microscope's resolution is about a micron. Therefore, if the clusters are lesser than the micron size then it cannot be distinguished by the optical microscope. Although, nanometer-size objects can be investigated by an electron microscope; it has a fatal disadvantage of an evacuated sample room. The solvent has to evaporate before observation and that limits the investigation as the drying process exceeds the aggregations of particles. Donselaar et al. [10] succeeded in observing clusters of submicron scale in the magnetic nanofluids in a frozen state by an electron microscope. However, from a more critical viewpoint, even these sub-micron clusters might be formed during the quenching process.

When one studies the magnetic colloidal particle behavior in the Magnetic nanofluids, it is necessary to know how strong the magnetic field is at the particle position. The so-called local magnetic field at the particle position, H_{loc}, is different from an external field, H. In addition to H, there are magnetic fields generated by the permanent magnetic moments of all other surrounded magnetic particles. If the colloidal particles are uniformly distributed, the problem is not so difficult, but the existence of clusters in the nanofluids together with the colloidal particles makes the problem more complicated. The local field, H_{loc}, inside the cluster should be much stronger than that at the rest of the system. However, as the cluster is micron size it is difficult to measure H_{loc} with ordinary

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http://dx.doi.org/10.1016/j.ultras.2014.08.017
0041-624X/

equipment however, such measurements can be done with the nuclear magnetic resonance (NMR) method [11,12]. By NMR experiment, not only the local field of the magnetic nanofluids were measured, but also the magnetization and particle concentration in the cluster can be obtained.

The small-angle neutron scattering (SANS) is quite sensitive to the aggregation processes in magnetic fluids [13]. Wide possibilities of the contrast variation (hydrogen/deuterium isotopic substitution) in neutron experiments allow us to 'look' inside the aggregates. Also, the additional magnetic scattering of neutrons can be used for studying magnetic correlations in nano-systems with magnetic inclusions. SANS distinguishes between magnetic and nonmagnetic components of ferrofluids allowing density, composition, and magnetization profiles to be precisely determined.

Ultrasonic propagation in magnetic fluid is a simplest non-destructive method to investigate the structure formation without any prior modifications of the sample. Several studies have been performed to investigate the properties of ultrasonic propagation in magnetic fluid prepared in polar or non-polar carrier [14–21]. In order to use this method for velocity profile measurement, it is important to have an accurate measurement of sound velocity in a magnetic fluid.

In the present paper we report the variation in ultrasound velocity, υ, as a function of solid volume fraction for different temperature and magnetic field of magnetic fluids prepared in two different carriers. These carriers are widely used in many engineering devices. Using the results of ultrasonic velocity profile, rheological and density measurement, various acoustic parameters were derived to understand the effect of temperature and magnetic field. This helps to understand the mechanism of cluster formation and or interaction between particles in the magnetic fluids.

2. Experimental

Co-precipitation technique followed by digestion was used to prepare magnetic nanoparticles. The ratio of Fe^{2+} and Fe^{3+} was kept as 1:2. The particles were coated with oleic acid and then dispersed in kerosene and transformer oil [22]. The system is labeled as MFK and MFT respectively for kerosene base and transformer oil based fluid. The density of fluid was measured using specific gravity bottle of 10 ml capacity. The Bruker powder X-ray diffractometer model D2 Phaser with LYNEX EYE detector was used for the structural investigation of particles. The data were collected at 2θ angle from 10° to 80° with 0.02° steps. Philips Tecnai F20 was used to study the morphology of the particles. The magnetic properties of fluids were measured using Polytronic magnetometer model BCS-100 using the principle of extraction method.

The ultrasonic sound velocity in the fluids was measured using the continuous wave ultrasonic interferometer (Mittal Enterprises) working at 2 MHz frequency with the accuracy of ±2 m/s. A digital micrometer screw (least count 0.001 mm) is used to lower or raise the reflector plate connected to the cell. An experimental set-up is shown in Fig. 1. The specially designed jacketed measuring cell was used to maintain the uniform temperature of the sample. The inlet and outlet of the cell is connected to constant temperature bath with the accuracy of ±0.1 K. The measuring cell containing approximately 3 ml of the sample is connected to the frequency generator using co-axial wire. The generator was fixed at 2 MHz frequency. The readings were noted by moving the micrometer screw, when current meter shows maximum deflection. The measuring cell was placed between the pole pieces of an electromagnet. The direction of magnetic field is perpendicular to the direction of ultrasonic wave propagation. The data were taken after 20 min of the application of magnetic field so as the system reach to the equilibrium [23].

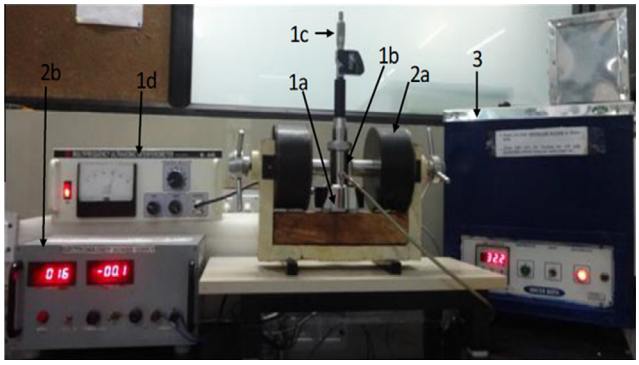

Fig. 1. Experimental set-up 1: (a) base to hold cell, (b) double jacketed measuring cell containing quartz crystal for generating 2 MHz frequency, (c) top part of the cell with micrometer screw gauge which moves reflector plate up and down and (d) multifrequency ultrasonic waves generator. 2: electromagnet setup (a) electromagnets, (b) DC power supply. 3: constant temperature bath.

Viscosity of the samples were measured using 18318 Rheolab QC (Anton Paar) attached with DG 26.7 measuring cup under constant shear rate (CSR) mode. Temperature was controlled with the accuracy of ±0.1 K using constant temperature bath.

3. Results and discussion

The X-ray diffraction pattern shown in Fig. 2 represents a single phase cubic spinel structure without any impurity phases. The broadness of all peaks indicates a typical characteristic of nanosize particles. Enhancements in intensity of peaks reveal the good crystallinity of the particles. Lattice parameter calculated from the pattern analysis is found as 0.8404 ± 0.0002 nm. This value is close to the bulk value for Fe_3O_4 system (0.8396 nm) [24]. The size of the particles calculated using Scherer's formula for the most intense (3 1 1) reflection plane is 11.5 nm. The morphology of the particles as seen from TEM image shows spherical shape particles. The particle diameter was measured from the different portion of the image and then plotted in histogram. The distribution of particle size thus observed is fitted with the log-normal diameter distribution function as described in Eq. (1). From the fit, the particle size is found as 11.6 nm with size distribution, σ as 0.25.

$$f(D)d(D) = \frac{1}{\sqrt{2}\pi\sigma D}\exp\left(\frac{-\ln(D/D_m)^2}{2\sigma^2}\right)dD \quad (1)$$

Here, $f(D)d(D)$ is the log-normal diameter distribution function with median diameter D_m and σ is size distribution in $\ln(D)$. The solid volume fraction of the particles was determined using the actual density of carrier ρ_c, density of particles, ρ_p (5 g/cc) and the density of the fluid, ρ_f. The formula used to calculate the solid volume fraction from the density is given in Eq. (2). Total five samples in kerosene and four samples in transformer oil were prepared with different volume fractions. The solid volume fraction of these samples is reported in Table 1. All the figures were drawn by considering the solid volume fraction of the particles.

$$\varphi = \frac{(\rho_f - \rho_c)}{(\rho_p - \rho_c)} \quad (2)$$

The magnetic measurement of all fluid samples was investigated using extraction method. Fig. 3 shows the magnetic response of the fluid under the influence of magnetic field. The fluid magnetization of samples was calculated using fluid density and quantity of sample taken for the measurement. It is seen that as the volume fraction of the fluid increases the magnetization increases from 116 to 312 kA/m for transformer oil based fluid while for