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Ultrasonic propagation: A technique to reveal field induced structures in magnetic nanofluids

Kinnari Parekh [a,*], Jaykumar Patel [a], R.V. Upadhyay [a,b]

[a] Dr. K C Patel R & D Center, Charotar University of Science & Technology, Changa 388 421, Dist. Anand, Gujarat, India
[b] P D Patel Institute of Applied Sciences, Charotar University of Science & Technology, Changa 388 421, Dist. Anand, Gujarat, India

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ABSTRACT

The paper reports the study of magnetic field induced structures in magnetic nanofluid investigated through ultrasonic wave propagation. Modified Tarapov's theory is used to study variation in velocity anisotropy with magnetic field. The types of field induced structures depend upon the chemical structure of the carrier in which magnetic nanoparticles are dispersed. Our study indicates formation of fractals and chain respectively, in transformer oil and kerosene based fluid. This difference is explained on the basis of particle–particle interaction and particle–medium interaction.

1. Introduction

Ultrasonic propagation in magnetic fluid is a simplest non-destructive method to investigate the structure formation without any prior modifications of the sample. Several experimental and theoretical studies have been performed to investigate the properties of ultrasonic propagation in magnetic fluid prepared in polar or non-polar carrier [1–8]. It seems that the spatial ordering of magnetic nanoparticles influences the ultrasonic propagation and by analyzing this it is possible to understand the homogeneity of magnetic fluids. 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 absence of external magnetic field, magnetic fluid behaves like a normal suspension containing magnetic nanoparticles. Under the influence of external magnetic field, the dipole moment is induced in the particles as a result, particles experience a Zeeman force and respond to the magnetic field either through Neel rotation or through Brownian rotation. In addition to the particle–field interaction, particles experience short range van der Waals and magnetic dipolar attractive forces. The collective behavior of the same will produce the structures within the carrier matrix. Parameters that influence the ultrasonic velocity anisotropy comprises of external parameters (magnetic field strength and its direction, temperature, frequency, measurement time scale) to microstructural parameters like types of particles, carrier, volume fraction, size, size distribution). Experimental results of aggregation and structuring of dispersed ferromagnetic particles in magnetic fluid have been explained theoretically by Tarapov et al. [9] using an ultrasound wavelength of the order of 10^{-4} m which is much greater than the dimension of the aggregates ($\sim 10^{-6}$ m). For such a high wavelength, the fluid behaves as if it is a homogeneous fluid, but the presence of aggregation can change the equation of magnetic state and the thermodynamic properties of magnetic fluid making them anisotropic and inhomogeneous. This will affect the collective behavior of magnetic fluid in the presence of a magnetic field.

The inhomogeneity of the fluid will modify the flow behavior as well as thermal transport properties needed for the heat transfer devices. Understanding the formation of structures is very important as it helps to develop some devices such as optical modulators, gratings, photonic devices [10,11], heat transfer devices with tunable thermal conductivity [12], dampers, speakers and seal with tunable viscosity [13] and many more. The association of ferromagnetic particles with or without magnetic field has been a subject of interest since more than six decades and is theoretically explained using various approximation models [14–16].

In our earlier work, the ultrasonic properties of magnetic fluid have been investigated as a function of particle concentration, temperature and magnetic field [17]. Under magnetic field, the attenuation of ultrasonic wave was found to decrease in transformer oil based fluid while in a kerosene base fluid it remains almost constant. With increasing concentrations, attenuation increases, but with magnetic field the trend remains same. Since the attenuation

* Corresponding author.
E-mail address: kinnariparekh.rnd@ecchanga.ac.in (K. Parekh).

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of ultrasonic wave is sensitive to the homogeneity of the medium, the results were explained based on the possibility of structure formation in the system. The decrease in attenuation at higher field strength in transformer oil based fluid can be correlated with the increase in the structure formation. While in kerosene base fluids such change in attenuation is not observed. Moreover, the results could not predict the types of structure formation in the medium.

The velocity anisotropy as a function of magnetic field as described by Tarapov et al. [9] can be used to retrieve such information's in magnetic fluid. In the present paper we report the detail analysis of velocity anisotropy versus magnetic field for the Fe_3O_4 fluids prepared in kerosene and transformer oil with variable concentration and temperature. The choice of these carriers is because of its wide use in many engineering devices. Results are analyzed using the concept of particle–particle interaction and particle–field interaction which helps to understand the role of carrier and concentration on the types of structure formation under the influence of magnetic field.

2. Experimental

Co-precipitation technique followed by digestion was used to prepare magnetic nanoparticles. The ratio of Fe^{2+} and Fe^{3+} was kept at 1:2. The particles were coated with oleic acid and then dispersed in kerosene and transformer oil [18]. The system is labeled as MFK and MFT respectively, for kerosene base and transformer oil based fluid.

The powder X-ray diffraction pattern measured using Bruker D2 Phaser with LYNEX EYE detector shows single phase spinel structure with 11.5 nm crystallite size. The morphology of the particles as seen from Philips Tecnai F20 TEM image shows spherical shape particles. The particle diameter distribution was fitted with the log-normal diameter distribution function which shows a median size of 11.6 nm with size distribution, σ as 0.25 [17].

The room temperature magnetic properties of fluids were measured using Polytronic magnetometer model BCS-100. The magnetic measurement of all fluid samples was investigated using extraction method. A typical magnetic response of the fluid under the influence of magnetic field (H) for MFK4 and MFT4 is shown in Fig. 1. In the absence of the magnetic field the tiny magnetic particles are moving randomly with its magnetic axis pointing in all directions. As a result, the net magnetization of the fluid is zero. Under the influence of magnetic field, the moments of the particles align in the field direction through Neel rotation or Brownian rotation. This alignment increases with increasing field strength resulting in incremental field response. The nature of this response is decided by the collective effect of the magnetic moment of particles, particle size, size distribution and inter particle interaction. For non-interacting system, the fluid magnetization (M) can be fitted with the Langevin's theory modified for moment distribution as illustrated in Eq. (1).

$$\begin{aligned} M(H,T) &= n\int_{\mu_{\min}}^{\mu_{\max}} \mu L(\xi)P(\mu)d\mu \\ &= n\int_{\mu_{\min}}^{\mu_{\max}} \mu\left\{Coth\left(\frac{\mu H}{k_BT}\right) - \frac{k_BT}{\mu H}\right\}\left(\frac{1}{\sqrt{2\pi}\sigma_\mu \mu}\right)\exp\left(\frac{\ln\left(\frac{\mu}{\mu_0}\right)^2}{2\sigma_\mu^2}\right)d\mu \end{aligned} \quad (1)$$

Here, n is number density of the particles, $L(\xi)$ is Langevin's function with ξ is a Langevin parameter ($=\mu H/k_BT$). $P(\mu)d\mu$ is the log-normal moment distribution function, μ represent the particle magnetic moment, μ_0 represent mean magnetic moment and σ_μ represent distribution in $\ln(\mu)$. k_B is the Boltzmann constant and T is the absolute temperature.

In Fig. 1a, the symbols are experimental data which are fitted with Eq. (1). The fit parameters viz; magnetic moment, moment distribution and saturation magnetization of fluid was deduced for all samples and is reported in Table 1. The fit shows the high value of magnetic moment of the transformer oil based fluid compared to that of kerosene based fluid (inset Fig. 1a). However, the moment distribution remains almost identical. The distribution of magnetic moment for both the system is illustrated in Fig. 1b. It is seen that for MFT system the curve is shifted towards large moment compared to that of MFK system. This will result in the higher value of initial susceptibility in transformer oil based fluid compared to kerosene based fluid. Since the same particles are dispersed in two different mediums, it is easy to correlate the higher susceptibility value with the possibility of cluster formation in transformer oil. The compatibility of oleic acid with kerosene and transformer oil supports this possibility. Inset Fig. 1b shows plot of the saturation magnetization versus volume fraction. The variation is linear for both kerosene as well as transformer oil based fluid confirming that fluid is insensitive to dilution. This also means that the clusters were formed in the initial stage only and then with dilution it is not broken. The dipole–dipole interaction parameter, λ, is calculated using the fit parameter which comes out to be 2.3 and 2.7, respectively for MFK and MFT system at 308 K temperature.

The ultrasonic 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. The specially designed jacketed measuring cell was used to maintain the uniform temperature of the sample. The inlet and outlet of the cell are connected to a constant temperature bath with the accuracy of

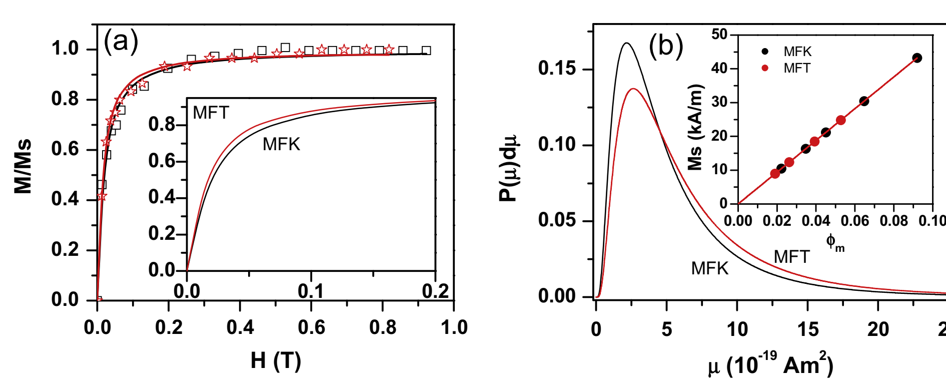

Fig. 1. (a) Magnetic response of kerosene and transformer oil based fluid under the influence of magnetic field. Line through data points represent Langevin fit using Eq. (1). (b) Moment distribution for MFK and MFT system. The inset figure shows the variation of fluid magnetization as a function of the different magnetic volume fraction.