

MAGNETIC PROPERTIES AND ANISOTROPY OF ULTRASOUND ATTENUATION IN APG-832 MAGNETIC LIQUID

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The results of the ultrasonic and magnetic investigations of APG-832 magnetic liquid are presented. The magnetic measurements were carried out with the aid of a vibrating sample magnetometer (VSM). On the basis of the magnetization curve, the coercive field strength, saturation magnetization and initial susceptibility were evaluated. The attenuation of ultrasonic wave in an APG832 magnetic fluid was studied using the broadband ultrasonic spectroscopy. In the external magnetic field the anisotropy of a sound attenuation was observed. This effect is attributed to the translational and rotational motions of the clusters of the colloidal magnetic particles dispersed in the carrier fluid.

Keywords: magnetic fluids, acoustic spectroscopy, VSM, magnetization.

1. Introduction

Ferrofluids are stable colloidal suspensions of nano-sized magnetic particles in a carrier liquid medium such as water or hydrocarbon. Their stability is achieved by coating them with the surfactant layer that produces entropic repulsion. However, the break in the balance between the Van der Waals attractive forces and the steric repulsive forces or the application of strong external magnetic field, causes certain amount of colloidal particles to join into spherical and chain-like aggregates (clusters) as long as hundreds of nanometers or more [1].

An external magnetic field acting on a magnetic liquid gives rise to a net magnetization of the sample that achieves saturation at a well-defined value of magnetic field strength. The physical mechanism underlying the macroscopic magnetization of a magnetic liquid in an external magnetic field, is the rotational degree of freedom exhibited by the magnetic particles. Thus, the magnetic field is able to orient the particle magnetic dipoles along the field direction.

A useful way to study the properties of magnetic liquids is based on application of ultrasonic methods [2]. They can be used to study the evolution of the structure

of magnetic liquid in variable magnetic and thermodynamic conditions. The structural changes are manifested by dependence of the ultrasonic wave absorption and velocity on the magnetic field intensity, direction of the wave with respect to the magnetic field and frequency of the wave.

2. Magnetic properties of APG-832 magnetic liquid

Magnetic measurements were carried out in a magnetic liquid denoted APG-832 (provided by Ferrotec Corp.), containing magnetite particles in a synthetic hydrocarbon. The magnetization curve was obtained with the aid of a vibrating sample magnetometer (VSM). The resulting hysteresis loop is shown in Fig. 1. With the increase of the applied magnetic field, magnetization of the sample reaches its saturation value of $M_s = 13.72 \text{ kA m}^{-1}$. In the saturated state all magnetic moments are oriented in the field direction. From the saturation magnetization M_s , the volume fraction of magnetite phase can be determined by means of the expression

$$\phi_V = \frac{M_s}{M_{\text{grain}}}, \quad (1)$$

where $M_{\text{grain}} = 446 \text{ kA/m}$ is the spontaneous magnetization of magnetite grains. In case of the APG-832 magnetic liquid, Eq. (1) leads to a value of 3%.

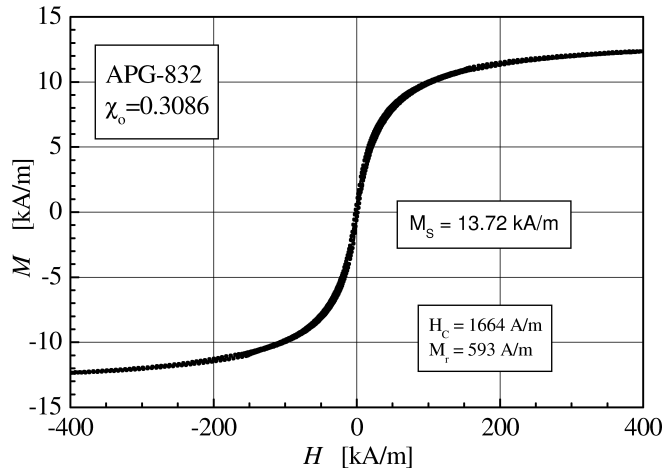


Fig. 1. Hysteresis loop for APG-832 magnetic liquid.

The process of magnetization and demagnetization of the ferromagnetic material is accompanied by energy loss at the expense of the external magnetic field, leading for instance to the increase of the temperature of the sample. Magnetic liquids, in general, exhibit superparamagnetic behavior with zero coercivity and remanence (no hysteresis loop). However, from Fig. 1 it is seen that APG-832 magnetic liquid shows small coercive force of $H_c = 1664 \text{ A m}^{-1}$. This proves that the fraction of magnetic grains reveals, aside from superparamagnetic, also partly ferromagnetic properties.

3. Ultrasonic anisotropy in APG-832 magnetic liquid

In the ultrasonic measurements, the broadband transducer (Optel) with center frequency of 5 MHz was used. The transducer was driven by MATEC Pulsar/Receiver SR-9000 Card, which provides unipolar spike pulse with amplitude of 400 V and rise time of 8 nanoseconds. The received signal was sampled and recorded in a digital oscilloscope LeCroy 9310AM. The power spectrum of the echo signal was determined by using a Fast Fourier Transform algorithm, with the aid of MatLab 9.0 software package. The measuring cell (made of brass) with fixed distance between the transducer and reflector was used. The path length (back and forth) traversed by the ultrasonic pulse inside the medium was 12 mm.

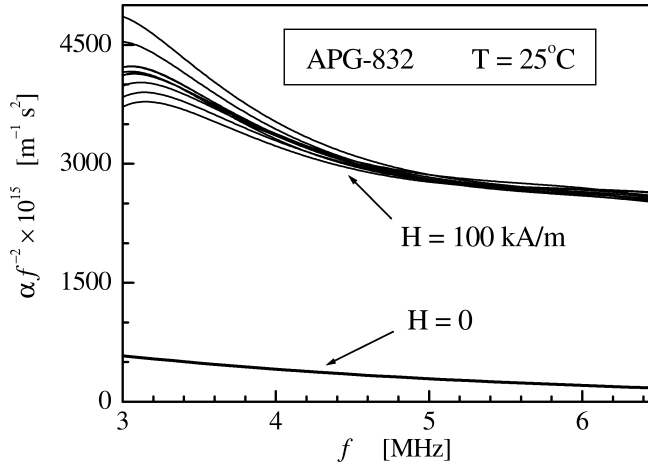


Fig. 2. Experimental results of the frequency dependence of ultrasonic attenuation in the magnetic liquid APG-832 at temperature 25°C, with and without external magnetic field. The curves for magnetic field equal to 100 kA m⁻¹ represent attenuation for different angles between the magnetic field lines and the direction of ultrasonic wave, from down to up: 90°, 30°, 40°, 0°, 60°, 80°, 10°, 50°, 70°, and 20°.

The attenuation coefficient measured in excess to that of the reference medium (water), can be expressed as [3]

$$\alpha(f) = -\frac{1}{2L} \ln \left(\frac{P_m(f)}{P_w(f)} \right) + \frac{1}{2L} \ln \left(\frac{R_{mb}}{R_{wb}} \right), \quad (2)$$

where $P_m(f)$, $P_w(f)$ are the power spectrums of the echo signal reflected from the wall of a measuring cell filled with magnetic liquid and water, respectively, $R_{mb} = 0.8335$ is the acoustic power reflection coefficient at the inner side of the measuring cell containing magnetic liquid, and $R_{wb} = 0.8314$ is the acoustic power reflection coefficient at the inner side of the measuring cell containing water. The accuracy of the ultrasonic measurements described above amounted to about $\pm 2-5\%$. For the angular dependence experiment, the magnetic field was rotated by ten degrees each time while the measuring cell remained stationary in the gap between pole pieces of electromagnet,

which yielded a field of 100 kA m^{-1} . The magnetic field strength was measured with a Resonance Technology RX21-type teslameter with the accuracy of 0.5%.

Figure 2 shows the experimental results of the attenuation per frequency squared, over the frequency range from 3 to 6 MHz, at temperature 25°C , in the magnetic liquid APG-832. As it is seen from the figure, in absence of the external magnetic field α/f^2 decreases monotonously and the attenuation can be attributed to the internal friction and heat exchange between the particles and the surrounding medium [4]. Application of the magnetic field to the fluid causes a substantial increase of the coefficient of attenuation due to the magnetoviscous effect [5]. Additionally, anisotropy in the attenuation of sound appears. The anisotropic contribution to the attenuation depends on the frequency of the wave and increases with the decrease in the frequency. The maxima on the attenuation curves, visible in the low frequency region, can be explained by oscillation of the chain-like clusters which tend to align with the field direction. If the frequency of the wave match the eigenfrequency of the internal chain vibrations, the attenuation will reach a maximum value. The angular dependence of the sound wave attenuation, for different frequencies in APG-832 magnetic liquid, is shown in Fig. 3.

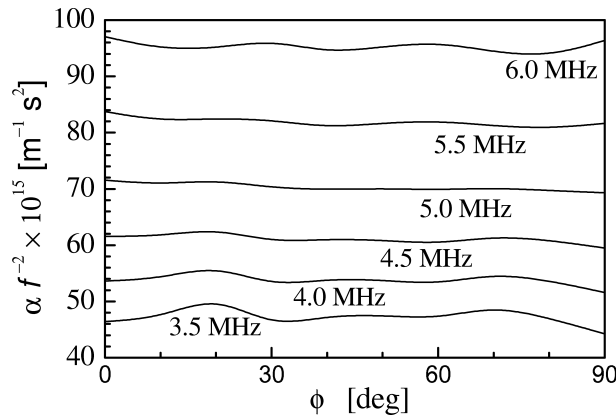


Fig. 3. Angular dependence of ultrasonic attenuation in the magnetic liquid APG-832 for different frequencies, in the external magnetic field of $H = 100 \text{ kA m}^{-1}$.

The results of the frequency dependence of ultrasonic attenuation show that the ferrofluid sample subjected to an external magnetic field indicate a formation of chains arranged along the lines of the external field. The chains are composed of spherical clusters whose size can be estimated by the ultrasonic measurements. According to TAKETOMI [6], the energy of the propagating ultrasonic waves in ferrofluid subject to an external magnetic field (showing anisotropy of elastic and magnetic properties), is used for activation of the translational and rotational degrees of freedom. Therefore the attenuation coefficient $\alpha(\theta)$ of an ultrasonic wave in ferrofluids subjected to an external magnetic field consists of two parts related, respectively, to translational motions of the clusters, $\alpha_{\text{tr}}(\theta)$, and their rotational motions, $\alpha_{\text{rot}}(\theta)$:

$$\alpha_{\text{tr}}(\theta) = \frac{1}{c} \frac{3\pi\eta_S r_{cl}\omega^3 V_{cl} N (6\pi\eta_S + V_{cl}\omega\rho_0)}{(k \sin \theta - V_{cl}\omega^2 \rho_m)^2 + (6\pi r_{cl}\omega\eta_S)^2}, \quad (3)$$

$$\alpha_{\text{rot}}(\theta) = \frac{\omega^2}{2\rho_0 c^3} \left(\frac{4}{3}\eta_S + \eta_V + 2\alpha_5 \cos^2 \theta + \alpha_1 \cos^4 \theta \right), \quad (4)$$

where c is the velocity of the ultrasonic wave propagating with angular frequency ω , ρ_0 and ρ_m are the densities of the magnetic liquid and magnetite grains, η_S and η_V are the dynamic and volume viscosities, r_{cl} and V_{cl} are the radius and volume of the cluster, N is the number of clusters per unit volume, k is the elastic force constant, θ is the angle between the magnetic field strength vector and the propagation vector of the ultrasonic wave, and α_1 , α_5 are the Leslie coefficients appearing in the stress tensor of liquid crystals.

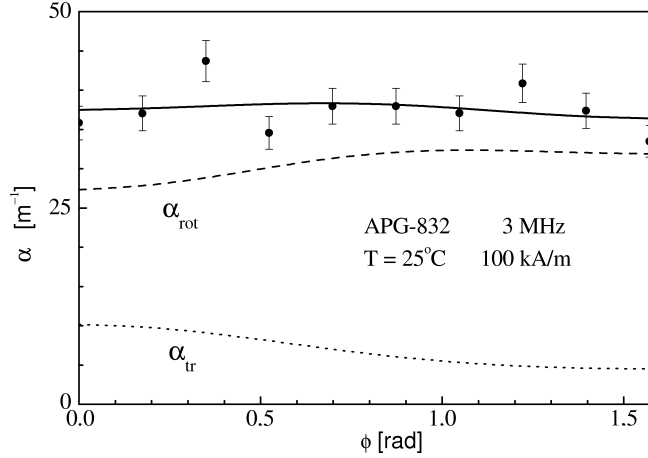


Fig. 4. Angular dependence of ultrasonic attenuation α for the wave of frequency $f = 3$ MHz in APG-832 magnetic liquid, in the external magnetic field of $H = 100$ kA m $^{-1}$. The solid line represents the sum of attenuation due to rotational motions of the clusters and translational motions of the cluster chains. Both contributions are also shown separately as the dashed and dotted lines, respectively.

Figure 4 shows the results of the ultrasonic anisotropy data $\alpha(\theta)$ for a APG-832 magnetic liquid subjected to a constant magnetic field of $H_{DC} = 100$ kA m $^{-1}$. The solid, dashed and dotted lines were obtained by fitting the sum of Eqs. (3) and (4) given in the Taketomi's theory to the experimental data. The fitting parameters, $4\eta_S/3 + \eta_V$, α_5 , α_1 , k , N , and r_{cl} , are listed in Table 1. The fitting procedure was carried out assuming $\eta_0 = 0.2$ Pa s, $c = 1313.3$ m s $^{-1}$, $\rho_0 = 1060$ kg m $^{-3}$, and $\rho_m = 5080$ kg m $^{-3}$.

Taking into account the concentration of magnetite particles and results of the fit (N and r_{cl} from Table 1), it is possible to estimate the percentage of the magnetic particles involved in the chain clusters from the equation:

$$\phi_{cl} = \frac{N4\pi r_{cl}^3}{3V_m} \times 100\%, \quad (5)$$

where V_m is the total volume occupied by all magnetic particles (both bounded in the clusters and flowing freely in a carrier liquid). The obtained percent of magnetic particles forming the chain clusters in the sample was 3.7%, so only a small fraction of

magnetic particles was engaged in forming the linear chains. This means that APG-832 magnetic liquid remains very stable in the presence of a strong magnetic field.

Table 1. Values of the parameters $4\eta_S/3 + \eta_V$, α_5 , α_1 , k , N and r_{cl} obtained by fitting the function describing the anisotropy of the ultrasonic absorption coefficient to the experimental points (at $T = 25^\circ\text{C}$; $H_{DC} = 100 \text{ kA m}^{-1}$), using the least-squares method.

| f [MHz] | $4\eta_S/3 + \eta_V$ [Pa s ⁻¹] | α_5 [Pa s ⁻¹] | α_1 [Pa s ⁻¹] | k [N m ⁻¹] | $10^{-15}N$ [m ⁻³] | $10^9 r_{cl}$ [m] |
|--------------|---|-------------------------------------|-------------------------------------|-----------------------------|-----------------------------------|----------------------|
| 3 | 0.427 | 0.0294 | -0.120 | 24.6 | 6.89 | 363 |
| 4 | 0.371 | 0.0048 | -0.037 | 52.4 | 2.08 | 472 |

4. Conclusions

The magnetic and ultrasonic measurements provide important information on the ferrofluid structure in a magnetic field. Analysis of the magnetization curve allowed us to determine the values of coercive field strength, saturation magnetization, and initial susceptibility. On the basis of the ultrasonic anisotropy, the main parameters describing the structure of a ferrofluid subjected to an external magnetic field of 100 kA m^{-1} were determined. Although a field of this strength caused the APG-832 magnetic liquid to reach a magnetization level which was close to the saturation value, still not all magnetic particles were involved in formation of the clusters. Majority of the particles were free in the carrier fluid.

Acknowledgments

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