Ultrasound Absorption in Magnetic Nanofluids

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ABSTRACT

A novel magnetodynamic mechanism of ultrasound absorption is proposed. It has relaxation form and reflects the specific properties of magnetic nanofluid due to the fact that nanoparticles of the solid phase have magnetic dipole moments. It is demonstrated that the most probable ultrasound energy dissipation channels are viscous, thermal and magnetodynamic. Comparison with existing experimental data is conducted. While the external magnetic field is applied the processes of nanoparticles aggregation are commenced. The aggregates appearing in magnetic nanofluid with external magnetic field applied are modeled by prolate spheroids of revolution. The analytical formula for ultrasound absorption coefficient in magnetic nanofluid with ellipsoidal particles is derived. The experimental results for ultrasound absorption anisotropy in dodecane-based magnetic nanofluid are received for the first time ever, and they demonstrate resonant ultrasound absorption at certain angle between ultrasound wave vector and direction of external magnetic field.

Keywords: magnetic nanofluid, ultrasound, absorption, anisotropy

1 MAGNETODYNAMIC MECHANISM OF ULTRASOUND ABSORBTION

Magnetic nanofluids are suspensions of very small ferromagnetic particles in carrier liquids. The diameter of these particles is usually 3 to 15 nm. Nanoparticle has a magnetic moment. Nanoparticles move in carrier liquid by random thermal motion. Brownian motion keeps nanoparticles from settling under gravity, and a surfactant is placed around each particle to provide short range steric repulsion between particles to prevent particle agglomeration [1].

The first results of studying magnetic fluids in the absence of external magnetic field by acoustic spectroscopy in the range of 3 to 50 MHz were presented in [2], [3]. Widening of the frequency range, in comparison to earlier works, made it possible to discover the relaxation character of ultrasound absorption. In [4], experimental results were given on measuring the ultrasound attenuation coefficient at seven frequencies in the range of 3 to 40 MHz in water-based magnetic nanofluid at three temperatures of 10, 20, and 30°C in the absence of external magnetic field. It was established that the ultrasound attenuation coefficient α/f^2 decreases nonlinearly with growth in frequency, and at a fixed frequency it decreases with increasing temperature. These data confirm the results given in [3] .

Micro-inhomogeneous media including magnetic nanofluid have two known channels of ultrasound energy dissipation: thermal and viscous [5] . Thermal channel is caused by equalization of temperatures on the boundaries of two different heated media under periodic changes of temperature, and viscous channel is caused by equalization of velocities due to different viscosity of the media It was shown that the viscous attenuation mechanism does not describe the experimental results for water, kerosene,and dodecane based magnetic nanofluids [6]. Here we have proposed a novel magnetodynamic mechanism of ultrasound energy dissipation in magnetic nanofluid, based on the fact that particles of magnetic nanofluid solid phase have their own magnetic moments. Ultrasonic wave with a frequency ω propagating in magnetic nanofluid with volume concentration φ and density ρ cause magnetic dipoles to oscillate thus exciting magnetic field of the same frequency and with amplitude h_0. Since the imaginary part of the magnetic susceptibility of the magnetic nanofluid χ'' is nonzero, then the energy of an alternating magnetic field is converted into heat, while the average dissipated power is determined, in accordance with the fluctuation-dissipation theorem, the expression

$$W = \frac{1}{2}\omega\chi''(\omega)\,{h_0}^2 \tag{1}$$

The attenuation coefficient of ultrasound in the general case is determined by the ratio

$$\alpha = \frac{W}{2c\overline{E}}, \tag{2}$$

where

$$\overline{E} = \frac{1}{2}\rho u_0^2\omega^2 \tag{3}$$

is the average flow of energy in the ultrasonic wave propagating in a magnetic nanofluid with a velocity c, u_0 is displacement amplitude of particles in the ultrasonic wave. Substituting eq.(1), eq.(3) in eq.(2), we obtain

$$\alpha_m = \frac{\chi''}{\rho c \omega}\left(\frac{h_0^2}{u_0^2}\right) \tag{4}$$

We have to estimate the ratio of the intensity of the local magnetic field to the amplitude of the displacement of the particles in the ultrasonic wave. To do this, we consider the change in the magnetic field on selected i-th dipole, so the corresponding displacement by a sound wave is equal to

$$u_i = u_0 \sin(\omega t - k r_i)$$

Consequently, due to the ultrasound wave changes the relative distance between the i- th and j - th particles is determined by

$$u_{ij} = u_0 k r_{ji} \cos(\omega t - k r_i)$$

Assuming spherical particles with a radius R uniformly distributed over the volume of magnetic fluid, whereas the average distance between particles r_m is equal

$$r_m = \left(\frac{4\pi}{3\varphi}\right)^{\frac{1}{3}} R$$

Changing the intensity of the local magnetic field caused by vibrations of particles in the ultrasonic wave, is given by

$$h_i = \frac{3 p u_0 k}{r_s^3}\cos(\omega t - k r_i).$$

Given the definition of the dipole moment of single-domain particle $p = {}^4\!/_3 \pi R^3 M_s$, and (4),we can derive the following expression for the attenuation coefficient

$$\alpha_m = \frac{9\chi''\varphi^2 M_s^2 \omega}{\rho c^3} \tag{5}$$

We denote the effective time of relaxation τ_m in a system of magnetic dipoles, and using the relationship of the real and the imaginary part of the magnetic field of the magnetic susceptibility [7].

$$\chi'' = \frac{\chi' \omega \tau_m}{1 + \omega^2 \tau_m^2},$$

and from eq.(5) we have obtained the final expression for the attenuation coefficient of ultrasound due to the magnetodynamic mechanism

$$\alpha_m = \frac{9\chi'\varphi^2 M_s^2 \tau_m \omega^2}{\rho c^3 (1 + \omega^2 \tau_m^2)}. \tag{6}$$

Experimental data on the frequency dependence of the additional attenuation of ultrasound satisfactorily described by the three mechanisms of energy dissipation of ultrasound: the viscous, magnetodynamic, and thermal, i.e.

$$\frac{\Delta\alpha}{f^2} = \left(\frac{\alpha}{f^2}\right)_V + \left(\frac{\alpha}{f^2}\right)_m + \left(\frac{\alpha}{f^2}\right)_{th}. \tag{7}$$

The contribution of viscous mechanism was estimated using the relation obtained in [8]:

$$\left(\frac{\alpha}{f^2}\right)_V = B\frac{\xi\left(1+\sqrt{\xi}\right)}{\left(1+\sqrt{\xi}\right)^2 + \xi\left(1 + b\sqrt{\xi}\right)^2}, \tag{8}$$

where

$$B = \frac{4\pi\varphi}{9c_f}\left(\frac{\rho_p}{\rho_f} - 1\right)^2, b = \frac{2}{9}\left(1 + 2\frac{\rho_p}{\rho_f}\right).$$

Here $\sqrt{\xi}$ the ratio of particle radius to the depth of penetration of the viscous wave.The radius of the particles R is variable parameter. Calculation of ultrasound attenuation due magnetodynamic mechanism was performed using (6), rewritten in the form

$$\left(\frac{\alpha}{f^2}\right)_m = \frac{A_R}{1 + \left({}^f\!/_{f_r}\right)^2}, \tag{9}$$

where f_r is the relaxation frequency in the magnetic subsystem. The attenuation of ultrasound due the thermal mechanism was evaluated using the expression obtained in [9]:

$$\left(\frac{\alpha}{f^2}\right)_{th} = \frac{2\pi^2 T c_0^2 \rho_f \rho_p C_{P_p}^2}{3ck_p} D\left(\frac{\alpha_p}{\rho_p C_{P_p}} - \frac{\alpha_f}{\rho_f C_{P_f}}\right)^2 \tag{10}$$

where

$$D = \varphi\left(\frac{k_p}{k_f} + \frac{1}{5}\right)R^2,$$

k_i is the thermal conductivity, C_{P_i} is the specific heat, α_i is the thermal expansivity In relations (7), (8) and (10) the index "p" refers to the values of the solid phase, and the index "f" - the carrier liquid. Equation (10) concludes that contribution of the thermal mechanism to additional absorbtion of ultrasound is independent of frequency, so this mechanism taken into account causes the reduction of the solid phase particles size.Following physical value were used in the calculations: $C_{P_p} = 655\,J/kg \cdot K$, $\kappa_p = 59\,W/m \cdot K$, $\alpha_p = 11,4 \cdot 10^{-6}\,K^{-1}$, $C_{P_f} = 2000\,J/kg \cdot K$, $\kappa_f = 0,12\,W/m \cdot K$, $\alpha_f = 9,5 \cdot 10^{-4}\,K^{-1}$.

Results of calculations for the kerosene-based magnetic nanofluid and the experimental data at $T = 293$ K are shown on fig. 1.

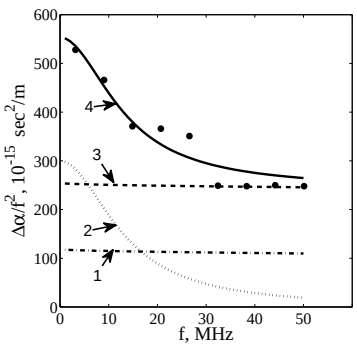

Fig.1 Additional absorbtion *versus* frequency.

Curve 1 describes the magnetodynamic mechanism, curve2 - the viscous mechanism, curve 3 - the sum of viscous and thermal mechanisms, curve of 4 - the additional attenuation calculated by eq.(7). The experimental data [2] are represented with small circles. The relaxation frequency in the magnetic subsystem was estimated about 12 MHz.

The results shown above are suitable for magnetic nanofluids with low concentrations. Analysis of acoustic spectra for magnetic nanofluid with high concentration of nanoparticles is much more difficult, since these fluids are non-Newtonian. The problems involve such aspects as non-additive contribution of different ultrasound energy dissipation mechanisms or absence of experimental results for shear viscosity frequency spectra. For instance, taking shear viscosity values for low-speed displacements yields in ultrasound absorption coefficient values much higher compared to experimental measurements.

2 ULTRASOUND ABSORPTION IN MAGNETIZED NANOFLUIDS

The processes of nanoparticles aggregation are commenced in amagnetic nanofluid when external magnetic field is applied. The aggregated are assumed to be the prolate ellipsoids and all have the same size at the fixed value of external magnetic field intensity. Then the dipole moment of such ellipsoid is expressed as $p = 4M_p\pi a^2 b/3$, where a and b are the semi-minor and semi-major axes of the ellipsoidal aggregate respectively, M_p is the saturation magnetization of solid phase substance in magnetic nanofluid. The magnetic field intensity is assumed to be great enough for the ellipsoids major axes to be oriented along its direction. The dipole repulsion forces between the adjacent ellipsoids only are taking into account with distance between them equaled to l, which value depends upon temperature, magnetic field intensity and solid phase concentration. The ultrasound wave propagation at angle ϑ to the direction of magnetic field causes ellipsoid displacement $\triangle x$, thereby the repulsive force of dipole-dipole interaction$F_d = -\kappa\triangle x \sin\vartheta$ tends to return the ellipsoid back to its equilibrium location. Force of friction affecting the oscillating ellipsoid with volume V_a in ultrasound wave is described with Stokes formula:

$$F = -\rho_f V_a \tau \frac{d\left(v_a - v_f\right)}{dt} - \rho_f V_a \omega S\left(v_a - v_f\right),$$

where

$$\tau = L + \frac{9}{4}\left(\delta_\eta/b\right)K^2,$$

$$S = \frac{9}{4}\left(\delta_\eta/b\right)K^2\left(1 + (1/K)\right)\left(\delta_\eta/a\right)$$

and "a" subscript denoting the properties of aggregates. Angular dependence of the inertial coefficient L in the expression for the added mass are defined due to symmetry as

$$L = L_\parallel \cos^2\vartheta + L_\perp \sin^2\vartheta\,,$$

$$K = K_\parallel \cos^2\vartheta + K_\perp \sin^2\vartheta.$$

where ϑ is the angle between the wave vector and ellipsoid major axis. The values $K_\parallel$,$K_\perp$ and $L_\parallel$,$L_\perp$ are defined according to [10]:

$$K_\perp = \frac{8}{3}\left(\frac{h}{h^2-1} + \frac{2h^2-3}{\left(h^2-1\right)^{\frac{3}{2}}}\ln\left(h + \sqrt{h^2-1}\right)\right)^{-1}$$

$$K_\parallel = \frac{8}{3}\left(-\frac{2h}{h^2-1} + \frac{2h^2-1}{\left(h^2-1\right)^{\frac{3}{2}}}\ln\frac{h+\sqrt{h^2-1}}{h-\sqrt{h^2-1}}\right)^{-1}$$

$$L_\parallel = \frac{\alpha_0}{2-\alpha_0}\,, \quad L_\perp = \frac{\beta_0}{2-\beta_0}\,,$$

$$\alpha_0 = \frac{2\left(1-e^2\right)}{e^3}\left(\frac{1}{2}\ln\frac{1+e}{1-e} - e\right)$$

$$\beta_0 = \frac{1}{e^2}\left(1 - \frac{1-e^2}{2e}\ln\frac{1+e}{1-e}\right), \qquad e = \sqrt{1-\frac{b^2}{a^2}}.$$

Ultrasound absorption coefficient in MNF is derived as

$$\alpha = \frac{1}{2}\varphi\frac{\omega}{c_f}\frac{\left(\frac{\rho_a}{\rho_f} - 1 - \frac{\kappa\sin\vartheta}{\omega^2\rho_f V_a}\right)^2}{\left(\frac{\rho_a}{\rho_f} + \tau - \frac{\kappa\sin\vartheta}{\omega^2\rho_f V_a}\right)^2 + S^2}S. \qquad (11)$$

It is important to note that in case of equal densities of aggregate and the carrier liquid eq.(11) is reduced to

$$\alpha = \frac{1}{2}\varphi\frac{\omega}{c_f}\frac{\left(\frac{\kappa\sin\vartheta}{\omega^2\rho_f V_a}\right)^2}{\left(1 + \tau - \frac{\kappa\sin\vartheta}{\omega^2\rho_f V_a}\right)^2 + S^2}S\,.$$

This absorption coefficient is caused by dipole interaction of adjacent aggregates only, and as a result, the

aggregates oscillation speed in the sound wave is different from the oscillating velocity of carrier liquid particles. Experimentally this situation can arise in magnetic emulsions. Another limiting case is $\kappa = 0$ when dipole-dipole interaction is absent, so eq.(11) is completely coincide with absorption coefficient formula derived in [11].

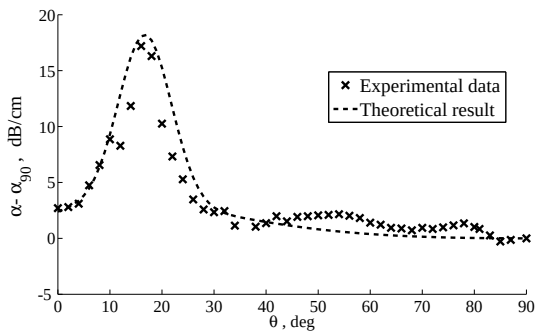

Fig. 2 Comparison of experimental and theoretical results.

The structural changes in a magnetic nanofluid at presence of external magnetic field are determined by diffusion processes, for the completion of which, as it turned out, sufficiently large time intervals are required. The specifics of a magnetic nanofluid were accounted for by the fact that the magnetic field was created with permanent magnets, since it had been experimentally established that the holding time of the magnetic nanofluid sample in the field, ensuring reproducibility of results, were tens and hundreds of hours. To obtain the angle dependencies of the acoustic properties, permanent magnets were set up on a rotating platform. Thermostating of the measuring cell was ensured with an accuracy of $\pm 0.05K$

Experimental results for ultrasound absorption anisotropy in dodecane-based magnetic nanofluid with volume concentration of magnetite particles of $\varphi = 0.1$ were obtained in thesis [12] and they are presented on Fig. 2. The dashed line is the computed result of eq.(11). Following physical value were used in the calculations:

$\rho_f = 749 kg/m^3$, $\rho_a = 4300\, kg/m^3$, $c_f = 1280\, m/s$, $\eta = 1.49210^{-3} Pa \cdot s$, $f = 3MHz$, $a = 2\, nm$, $l = 13.67 \cdot 10^{-8} m$.

Recently, there has been a tendency to study changes in ultrasound velocity and attenuation in non-equilibrium conditions due to overlapping of a magnetic field changing in time at a given rate. So, the authors of [13] measured the attenuation coefficient in a water-based magnetic nanofluid with a collinear and orthogonal magnetic field orientation with variation in the rate of magnetic field growth from $15 A/m \cdot s$ to $1.5 kA/m \cdot s$, and they obtained rather various field dependencies. However, in the majority of them, resonance ultrasound attenuation was observed.

3 CONCLUSION

Theoretical results of acoustic absorption spectra description are summarized in this paper. Novel magnetodynamic mechanism of ultrasound energy dissipation is proposed. The estimation of ultrasound absorption caused by this mechanism can be obtained with a different approach involving frequency dependencies of real and imaginary parts of magnetic susceptibility. However the experimental results of these dependences in megahertz-range are absent at present time. Analytical expressions for ultrasound absorption in media with ellipsoidal particles can be used in analysis of acoustic spectra of magnetorheological fluids.

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