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Effect of dilution on the water based starch coated magnetic fluid

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ABSTRACT

Ultrasonic study of the water based starch coated magnetite nano particles have been made with and without the external magnetic field at 303 K. The observations were recorded for six different volume concentrations of the same fluid under different magnetic field strength. The results show that the interactions between the solvent and the coated solute particles depend on the extent of applied field. The extent of external field increases the agglomeration and forms a chain structure that plays an important role in the determination of strength of grain-field interaction. Comparative study of the variation in acoustical properties suggests that the effect of dilution is to reduce the chain structure formation thereby weakens the effective interaction between the external field and the magnetic particles.

KEYWORDS

Ultrasonic velocity;
Ferro fluids;
Acoustic parameters;
Grain-field interactions.

INTRODUCTION

A magnetic fluid is a colloidal suspension of magnetic particles covered with a surface layer in a carrier liquid[1, 2]. Flow properties of even a normal colloid are some time unusual and distinctive[3], which in a ferrofluid additional complexity is introduced by subjecting it to an external magnetic field. Such a fluid exhibits pronounced non-Newtonian effects under the action of field[4].

Diluted particle dispersion is known to behave as a Newtonian flow and much research have been done on such systems[5]. Theoretical as well as experimental studies of these magnetically induced rheological effects are essential for designing certain magnetic fluidic devices and applications[6]. Under the influence of an external magnetic field, a ferrofluid reveals very interesting properties. In the presence of an external magnetic field, the magnetic particles join together forming chains or columns arranged along the direction of the field. Such process depends on the value of the magnetic field interaction[7]. The change in the ferrofluid structure affects the acoustic properties of the fluid such as the packing density, wave propagation velocity, etc. So ultrasonic methods can be used to investigate the structural changes in ferro fluids[7-9]. This paper reports the experimental synthesis of an aqueous based starch coated magnetite fluid and results of the study on the changes in ultrasonic wave velocity as a function of magnetic field, field orientation and dilution of carrier at 303 K.

SAMPLE PREPARATION

The studies were performed in the water-based ferrofluid containing magnetite nano particles. The particles are coated with starch, a long-chain polymer of D-glucose, which provides a good biocompatibility and is non-toxic. The fluid was prepared by the method of controlled co precipitation technique, which is given as follows.

Magnetite was precipitated by dissolving 2.08 g $FeCl_2$ anhydrous and 5.22 g $FeCl_3$ anhydrous in 380 ml of conductivity water and adding 20 ml of 25% NH_3 to this solution while stirring vigorously. After sedimenting the precipitate with a permanent magnet, the supernatant was removed by decantation. 40 ml of 2 M HNO_3 was then added to the black sediment and the mixture was stirred for 5 minutes. The oxidation of magnetite was completed by adding 60 ml 0.35 M $Fe(NO_3)_3$ to the mixture and stirring it at its boiling temperature for 1 hour. After sedimentation and washing with 2 M HNO_3, conductivity water was added to disperse the reddish yellow sediments. The resulting black dispersion contains 5.6 grams of solid material per litre.

Starch coating as surfactant to the particles is provided by the following procedure. 2 ml of the above product was diluted by adding 50 ml conductivity water. The solution was flocculated by adding few drops of 25% NH_3 and sedimented using a permanent magnet. After washing with 50 ml water, 100 ml water was added to the precipitate. Under mild mechanical stirring, 20 ml of the prepared starch solution was added. Within a few minutes, all magnetic material was transferred to the distinctive starch phase. The black oil droplets were separated from colorless water phase and washed many times with 10 ml ethanol to remove excess water and excess surfactant.

The basic properties of the ferrofluid such as the value of saturation field, shear viscosity, initial susceptibility, density and particle volume concentrations at 303 K are found at Ferrofluids Laboratory, Pondicherry, India and is known as 28 mT, 5.3 cp, 0.53, 1172 kg m^{-3} and 3.84 % respectively. Magnetic fluids of various volume concentrations were obtained from the initial one by dilution with water, adopting the usual dilution procedure[10]. All the samples were checked for their stability and reliability before recording of sound velocity data and they found to be quite satisfactory.

MEASUREMENT OF SOUND VELOCITY

The study was performed for six samples of the same ferrofluids containing different particle volume concentration of magnetite viz., 3.84 %, 3.46 %, 3.07 %, 2.67 %, 2.30 % and 1.92 %. They are indicated in the work as 0 %, 10 %, 20 %, 30 %, 40 % and 50 % of dilution respectively.

Velocity anisotropy was studied by using the variable path, fixed frequency, ultrasonic interferometer (Model F81 of Mittal Enterprises, India). The instrument generates a very stable ultrasonic frequency of 2 MHz (± 0.03%). The accuracy in the velocity is given as $\pm 0.01 ms^{-1}$. Details of the interferometer are described elsewhere[11, 12]. To measure the velocity at different magnetic fields, the measuring cell was inserted into an air core solenoid. Hibbert's magnetic standard was used to ensure the field inside the measuring cell to be of required value. Magnetic field strength was varied in steps of 500G from 0 to 5000G by varying current in the solenoid. It was possible to incline the solenoid so that the angle between the direction of the field and the direction of propagation of ultrasonic waves can be varied from 0 to 20° and can be set at 90°. As a ferrofluid need about 30 minutes to reach a new state of equilibrium[13, 14], each time when the field is changed the system is allowed for a minimum of 45 minutes so as to reach equilibrium and then the observations were made.

EVALUATION OF ACOUSTICAL PARAMETERS

Three of the acoustical parameters viz., the adiabatic compressibility (β), the intermolecular free length (L_f) and the acoustical impedance (Z) have been calculated using the standard relations[15, 16].

$$\beta = \left(\frac{1}{U2\rho} \right) \tag{1}$$

$$L_f = K_T \beta^{1/2} \tag{2}$$

$$Z = U \tag{3}$$

Where U is the ultrasonic velocity and ρ is the density and K_T is the temperature dependent constant. The value of K_T at 303 K is 199.53 x 10^{-8} in SI system.

RESULTS AND DISCUSSION

The experimental values of bulk density and ultrasonic velocity measured in the samples under no field conditions and the related acoustic parameters are presented in TABLE 1. TABLE 2 narrate the measured values of ultrasonic velocity for various field strength in two different inclinations (parallel and perpendicular). As the external field has no effect in bulk density of the sample, various acoustic properties have been calculated using the bulk density value measured under no filed conditions. The respective calculated values of adiabatic compressibility are presented in TABLE 3. The variations in the trend of free length & acoustic impedance are shown in Figures 1 to 4 respectively.

TABLE 1 : Measured and calculated parameters for the Ferrofluid Water + Magnetite + Starch at 303 K without external field

Dilution %	ρ kg m^{-3}	U ms^{-1}	? x 10^{10} Pa^{-1}	L$_f$ x 10^{11} M	Z × 10^{-6} kg m^{-2}s^{-1}
0	1160.2	1452.2	4.0871	4.0338	1.6848
10	1140.2	1454.6	4.1451	4.0623	1.6291
20	1118.7	1456.3	4.2149	4.0964	1.6291
30	1101.2	1457.9	4.2725	4.1243	1.6054
40	1086.2	1460.2	4.3178	4.1461	1.5861
50	1072.6	1462.3	4.3600	4.1663	1.5685

The perusal of TABLE 1 reveals that in the absence of external magnetic field, ultrasonic velocity is found to increase with dilution of fluid i.e., it approaches the velocity of sound in water. However, the density values are in decreasing trend. A non-linear variation in these parameters indicate that the medium is having appreciable degree of interactions[17-19] due to dilution but there would be no agglomeration or flocculation as there are no sudden abrupt changes. Visual observation of all the diluted samples reveals no precipitation / sedimentation. On dilution, the density of fluid gets reduced whereas the sound velocity is increased which implies that some form of compactness, exists between the components of the medium[20].

Any elevation in sound velocity may be attributed to two chances as (i) the increase in compactness of the medium or the reduction in free space between the components and (ii) an elevation in the pressure of the medium. For a given pressure and frequency as in this case it is only the compactness that enhances sound velocity. Compactness in turn, may be due to the development of size of the components or the increase in the number of component molecule. If the size of the component develops, more energy will be needed to overcome the inertial effects[21] and so this is overruled here. Thus the effect of dilution increases the number of components available in the medium and at the same time drastically increases the effective volume per unit mass and hence sound velocity increases whereas density decreases. However, the fluid is found to remain as magnetic fluid. It may due to the surfactant, which protects the nano particles from excess carrier, in doing so; it creates strong interaction between the carrier particles.

The calculated values of adiabatic compressibility and free length increase with dilution whereas the acoustic impedance decreases. The ease with which a medium be compressed is indicated by the compressibility values[22]. The free length is found to be a predominant factor in determining the nature of sound velocity variations in the liquid mixtures[23]. Thus the larger values of β with increasing percentage of dilution reveal that the coated particles are in a set away mood and thus an increase in distance of separation exits. This is reflected in the observed free length values. The extent of opposition offered to sound propagation is indicated by Z values. As the dilution is not only to increase the number of components in the medium but at the same time it replaces the surrounding atmosphere of the coated heavy particles by the light water particles, thereby reduces the inertial effects, acoustic propagation is made very easier or the repulsion to sound is reduced. Thus the Z values show a decreasing trend. The appreciable variation in the values with respect to the dilution suggests that the existing interactions are strong[24].

TABLE 2 presents the measured values of ultrasonic velocity in parallel and perpendicular directions for various external magnetic fields at 303 K. The trend of sound velocity is found to be increasing with respect to the field as well as dilution. For no dilution, velocity variations are less pronounced in parallel direction for lower filed values whereas it is highly pronounced than for perpendicular direction at higher field values. On dilution, the entire picture gets changed, as sound ve-

TABLE 2 : Measured values of velocity in ms^{-1}for various applied magnetic field at 303 K

Dilution %	Magnetic field strength (in Gauss)										
	Parallel field										
	0	**500**	**1000**	**1500**	**2000**	**2500**	**3000**	**3500**	**4000**	**4500**	**5000**
0	1452.2	1452.3	1452.5	1452.7	1453.1	1454.0	1454.5	1456.2	1457.3	1458.6	1459.5
10	1454.6	1452.4	1452.6	1452.9	1453.2	1454.6	1454.9	1456.3	1457.6	1458.9	1459.8
20	1456.3	1453.1	1453.6	1453.8	1454.0	1455.0	1455.2	1456.6	1458.2	1459.2	1460.2
30	1457.9	1453.8	1454.4	1454.6	1454.9	1455.4	1455.7	1456.9	1458.6	1459.9	1460.7
40	1460.2	1454.2	1454.9	1455.1	1455.3	1455.9	1456.2	1457.2	1459.2	1460.1	1461.6
50	1462.3	1455.1	1455.6	1455.9	1456.2	1456.8	1457.1	1457.8	1459.8	1461.2	1462.5
	Perpendicular field										
0	1452.2	1452.3	1452.8	1453.2	1453.7	1454.1	1454.7	1455.2	1456.0	1456.7	1457.5
10	1454.6	1452.4	1452.8	1453.2	1453.8	1454.4	1454.9	1455.3	1456.1	1456.8	1457.7
20	1456.3	1452.5	1453.0	1453.3	1454.0	1454.7	1455.2	1455.4	1456.4	1456.9	1457.9
30	1457.9	1452.6	1453.2	1453.5	1454.2	1455.0	1455.4	1455.7	1456.7	1457.2	1458.2
40	1460.2	1452.7	1453.6	1453.9	1454.6	1455.2	1455.8	1456.0	1457.0	1457.3	1458.6
50	1462.3	1452.8	1454.1	1454.4	1455.0	1455.6	1456.0	1456.3	1457.3	1457.9	1459.2

locity suddenly decreases with the onset of field for both orientations. However, it exhibits an increasing trend as external field increases. This sudden drop in velocity may be attributed to the chaotic repulsions offered by the diamagnetic nature of water[25]. As external field increases the existing ferro particles in the medium predominates the chaos of the diamagnetic career and hence sound velocity is in increasing trend in both orientations. For a given dilution, the increase in magnetic filed increases the sound velocity and in the similar way for a given field strength, the extent of dilution also increases the sound velocity. This is true for parallel as well as in perpendicular field. However, the variations are not much pronounced in the perpendicular field.

The increase in sound velocity with increasing magnetic filed is due to the reorientation of the magnetite particles along the direction of the filed. Depending on the direction and the strength of the external field, the orientation of magnetic diploes will be there. The continuous increase of velocity suggests that a chain like structure or cluster formation is possible between the magnetic particles. Thus it may lead to purely temporary agglomeration that cannot be avoided as suggested by[26]. However, for the perpendicular field, as the effective work done by the magnetic dipole will be zero[25], the velocity variations are not much pronounced. For the same reasons as cited already, the extent of dilution increases the sound velocity. It is peculiar to note that larger dilution and larger external field yields a maximum sound velocity that one may lead to think that highly diluted fluids can be suited for practical applications, which is not exactly so due to the drastic fall in their magnetic properties[27]. As regards the acoustic properties, the grain-field interactions are evident and found to be the deciding factor of the velocity in the medium. Thus the observation of this TABLE clearly reveals that the existing interactions are of grain-field type and the effect of dilution is unanimous in weakening the existing interactions and is independent of field strength and angle of inclination.

TABLE 3 present the values of β under various external fields whereas Figure 1 & Figure 2 depicts the variation of free length with external field. As expected they behave similar to each other and reverse to the sound velocity. The higher value of β at high percentage of dilution indicates the existence of the more amount of free space between the components. Thus the excess career are existing freely in between the coated particles[28] and hence L_f also shows an increasing trend.

With the onset of the external field in both orientations, for diluted cases, β shows a sudden increase. However the increasing external field tends to form many clusters, thereby reducing β. There is no such increase in β for undiluted sample, which clearly reveals that the dilution makes the medium to be more

TABLE 3 : Calculated values of adiabatic compressibility in 10^{-10} Pa^{-1} for various applied magnetic field at 303 K

Dilution %	Magnetic field strength (in Gauss)										
	Parallel field										
	0	500	1000	1500	2000	2500	3000	3500	4000	4500	5000
0	4.0871	4.0865	4.0854	4.0843	4.0820	4.0769	4.0742	4.0647	4.0585	4.0513	4.0463
10	4.1451	4.1576	4.1565	4.1548	4.1531	4.1451	4.1434	4.1354	4.1280	4.1207	4.1156
20	4.2149	4.2359	4.2305	4.2294	4.2282	4.2224	4.2212	4.2131	4.2039	4.1981	4.1924
30	4.2725	4.2966	4.2930	4.2918	4.2901	4.2872	4.2854	4.2783	4.2684	4.2608	4.2561
40	4.3178	4.3535	4.3493	4.3482	4.3469	4.3434	4.3416	4.3356	4.3237	4.3184	4.3096
50	4.3600	4.4033	4.4003	4.3984	4.3966	4.3930	4.3912	4.3869	4.3749	4.3666	4.3588
	Perpendicular Field										
0	4.0871	4.0865	4.0837	4.0814	4.0787	4.0764	4.0731	4.0703	4.0658	4.0619	4.0574
10	4.1451	4.1576	4.1553	4.1531	4.1496	4.1462	4.1434	4.1410	4.1365	4.1326	4.1275
20	4.2149	4.2349	4.2340	4.2323	4.2282	4.2242	4.2213	4.2201	4.2143	4.2114	4.2056
30	4.2725	4.3037	4.3001	4.2984	4.2942	4.2895	4.2871	4.2854	4.2795	4.2766	4.2707
40	4.3178	4.3625	4.3571	4.3553	4.3511	4.3475	4.3440	4.3428	4.3365	4.3350	4.3279
50	4.3600	4.4172	4.4093	4.4075	4.4039	4.4003	4.3978	4.3960	4.3899	4.3864	4.3786

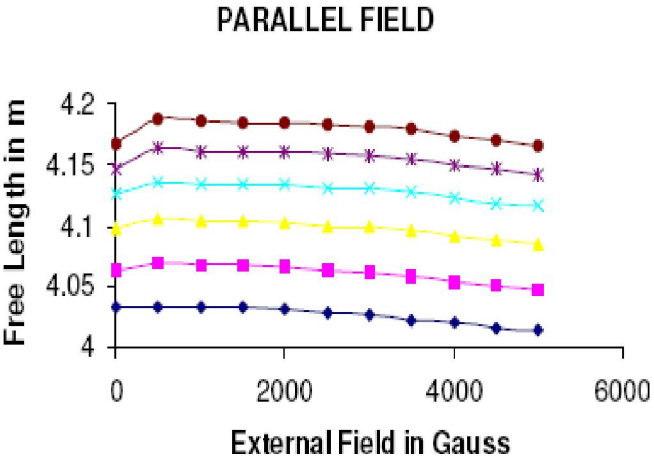

Figure 1 : Variation of Free Length with external field (Parallel) at 303 K.

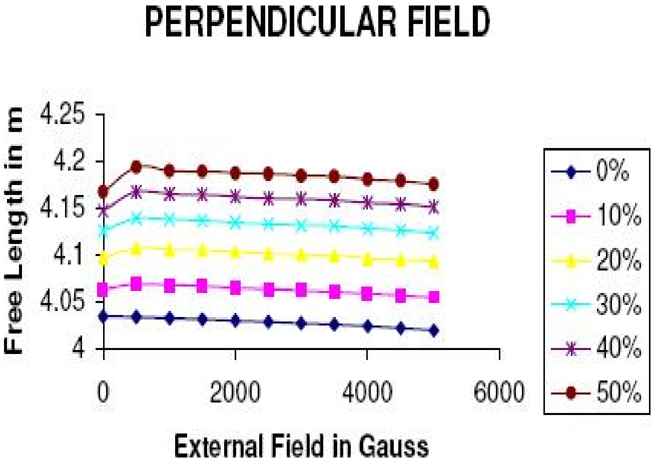

Figure 2 : Variation of Free Length with external field (Perpendicular) at 303 K.

rarefied of ferro particles, thereby weakening the existing interactions. Further the chaos between the diamagnetic career and the ferromagnetic particles are reflected in the sudden change with the onset of the field. As external field is increased the ferro magnetic particles, being solid, predominates the diamagnetic interruptions and hence cluster formation sustained at higher field values. The variations in the free length reflect similar suggestions of β. Such conclusions are supported by the findings of Taketomi[28] in which the effective cluster sizes have been detected.

The decrease in β and L_f with respect to the applied field indicates that the external field supports the chain structure formation. But the increasing trend of the parameters with dilution reveals that the formation of chain structure is not supported by dilution and further it leads to the indication of weakening of interactions as convinced by[29] in some sugar solutions. This is true for both parallel and perpendicular fields.

Z as is observed gives an exactly reverse trend to β and L_f from Figures 3 & 4. The weakening of interactions by dilution is again reflected here. i.e., in the case of diluted samples, Z exhibits a lower value if the external field is applied. In such cases, as excess career is available, the medium is more homogeneous for sound propagation Z decreases suddenly. However the onset of field makes the ferro particles to be highly dispersed and are interrupting the sound propagation, thus showing a depressed velocity associated with decreased Z value. However, the gradual alignment of the ferro particles by the increasing external field supports the acoustic transmission and hence the acoustic impedance increases.

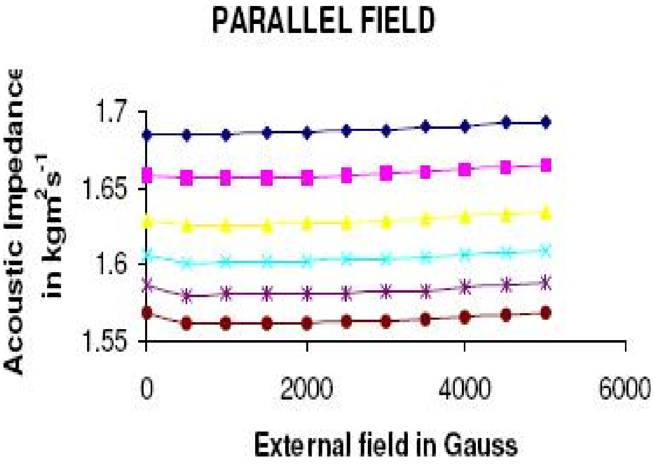

Figure 3 : Variation of Acoustic Impedance with external field (Parallel) at 303 K.

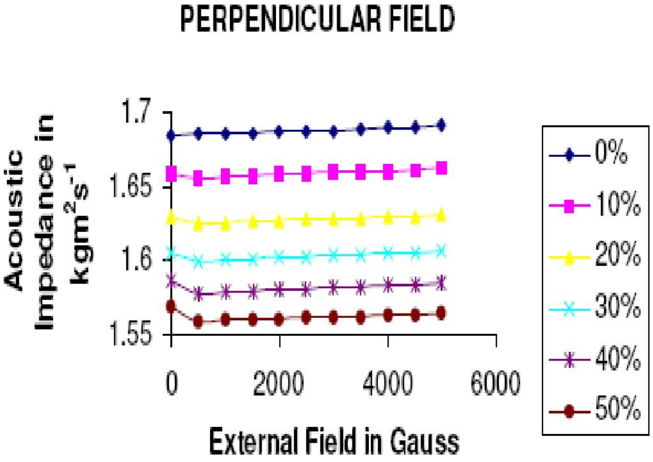

Figure 4 : Variation of Acoustic Impedance with external field (Perpendicular) at 303 K.

A continuous increase in Z with the applied field and a continuous decrease in Z with dilution are noticed. The clusters formed in the medium by the external magnetic field are elucidated and the sound energy that propagates has to overcome the heavy inertial effects of the formed structure. This is reflected in the enhanced Z value with respect to the field. The ultrasonic analysis in human cerebro spinal fluids done by[30] indicate that the increase in interactions are reflected by the enhanced Z values due to the elevated glucose level. Such observation forms a support for the present case in which the starch, the polymer of glucose in aqueous career can lend a support to the appreciable interactions. However as dilution increases the inertial effect is very much decreased, thereby pronounced decrease in acoustic impedance is noticed.

CONCLUSION

1. The existence of grain field interactions are confirmed.
2. The effect of dilution on the water based starch coated magnetite fluid is found to be reducing the chain structure formation and weakening the grain-field interaction.
3. The grain field interactions are evident at the enormous dilution also.

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