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The Effect of Temperature on the Viscosity of Cobalt Ferrite Nanofluids

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Abstract**:** ***The temperature dependence of viscosity is an essential property of the magnetic fluids applied for heat transfer systems. This property was considered in our work for Cobalt ferrite nanofluids - one of the most explored magnetic materials recently by their improved magnetic characteristics. Cobalt ferrite nanoparticles (CFNPs) were prepared by the co-precipitation method. The characterization of the synthesized particles was analyzed by various techniques such as X-ray diffraction, transmission electron microscopy, vibrating sample magnetometry, and thermal gravimetric analysis. The effect of temperature on the viscosity of Cobalt ferrite nanofluids was investigated. Experiments were carried out in the range of particle concentration from 0.5 to 7 % without and with a magnetic field application. The dynamic viscosities of these nanofluids were measured as the shear rate and temperature dependence under the magnetic field of different intensities, using a standard rotating rheometer. The cobalt ferrite fluids exhibit a yield shear-thinning behavior at all the temperatures from 25 to 55 ^{o}C. The experimental results show that the viscosity decreases when the temperature is increased. This variation is exponentially and dependent on the shear rate. The temperature-dependent viscosity is not influenced significantly by either particle concentration or magnetic field. From the obtained results, the Arrhenius equation for the viscosity-temperature relationship is applied.***

Keywords**:** ***Cobalt ferrite; Magnetic nanofluids; Viscosity; Temperature.***

I. INTRODUCTION

In recent years, magnetic fluids (MNFs) have gained the attention of the research community due to their potential use in technical and medical applications. The research focuses on the structure, dynamics of fluids, and the influences of corresponding parameters. Among that, the temperature is considered as the most critical parameter [1]. However, in most the fluid dynamic studies for MNFs, the viscous property of the fluid was assumed to be independent of temperature.

Recently a significant variation of the viscous property of MNFs due to change in temperature has been recorded in heat transfer systems [2], [3]. The temperature dependent viscosity becomes an extensive research field [2]-[8]. In general, the reports demonstrate a reduction in viscosity with increasing temperature. This variation, however, is quite complicated. Exponential correlations with different parameters were given in some works [2], [5], [8]. In contrast, a linear relation viscosity with temperature was reported for the ferrofluids at the temperature range of 20 - 50 ^{o}C [4], [7], meanwhile a no apparent change in relative viscosity as increasing temperature, was presented in [6]. It can be seen that almost researches were carried out for the nanofluids based on magnetite (Fe_3O_4) and maghemite (Fe_2O_3). Meanwhile, cobalt ferrite nanofluids (CFFs), considered as alternative materials that exhibit improved magnetic characteristics, such as unique optical, high saturation magnetization, strong magnetocrystalline anisotropy, and excellent chemical stability [9], has not been very well studied and understood As seen in the above studies, the viscosity-temperature dependence of CFFs needs to be investigated. Thus, in the present study, the effect of temperature on the viscosity of CFFs under an external magnetic field with various strengths was considered. The investigations were carried out for the CFFs with different particle concentrations.

II. EXPERIMENTAL

A. Materials

Ferric chloride hexahydrate (FeCl3.6H2O, AR) and Cobalt chloride (CoCl2.6H2O, AR) are provided by BDH Chemicals Ltd. England; Sodium hydroxide (NaOH) is supplied by Merck.

B. Cobalt ferrite fluid preparation

In this work, Cobalt ferrite nanoparticles (CFNPs) were prepared by the co-precipitation procedures as follows. The solutions of 1M $FeCl_3$ and 0.5M $CoCl_2$ both in 1M HCl were prepared as stock ones. The mixture of 20 ml of the stock solutions with the molar ratio of Fe^{3+} to Co^{2+} as 2:1 was added into a reaction vessel containing 100 ml of the NaOH solution, which was adjusted to the required pH of 12 and heated to the temperature of 30 ^{o}C. During the reaction time of 2 hours, the stir rate of 300 rpm and the temperature of 30 ^{o}C were remained. The reaction pH was kept unchanged by adding the 1.2 M NaOH solution continuously. The resulting precipitates were separated by a strong magnet and then washed many times with distilled water until the pH reached neutral and there was not chloride ion in the washed water.

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After that, the particles were dispersed in water at various weight fractions by ultrasonication for several hours until there was no visible precipitate

C. Characterization of Cobalt ferrite fluids

The crystalline structure of the prepared cobalt ferrite particles was characterized by X-ray diffraction (XRD) measurements, using a D8-AdvancBruker AXS diffractometer with CuKα radiation (λ = 1.542 Å) operating at 25 °C. The shape, size, and particle size distribution (PSD) of CFNPs were observed directly by transmission electron microscopy (TEM) technique with JEOL EM 1010. The magnetic characteristics, such as saturation magnetization and coercivity value, of CFNPs were evaluated by a Vibrating sample magnetometer (VSM) MicroSense EZ9. The VMS measurements were carried out from 0 to 16 kOe field and vice versa.

Nanoparticles weight fraction was estimated by a SETARAM thermogravimetric analyzer (TGA). The measurements were taken under nitrogen gas with a heating rate of 15 °C/min from 25 to 600 °C.

D. Viscosity measurement

A HAAKE RotoVisco 1 Rheometer was used to measure the viscosity of cobalt ferrite nanofluids, either the presence or the absence of a magnetic field. The measurement temperature of the fluids was controlled in the range 25 to 55 ± 1 °C by a thermostatic chamber. Since there was no precipitate observed in the fluid samples in stored days, the effect of sedimentation of the CFFs could be ignored in the rheological measurements.

III. RESULT AND DISCUSSION

A. Cobalt ferrite fluid characterization

The characteristics of the prepared particles and corresponding fluids are given in Table I. The XRD patterns of the samples are presented in Fig. 1. At all the samples, the XRD patterns (Fig. 1) of the synthesized nanoparticles show the characteristic peaks (220), (311), (400), (422), (511) and (440) which are matched to the cubic spinel structure (JCPDS card no. 22-1086). The average particle size (d_{avg}) and particle size distribution (PSD) of the particles were measured from TEM images (Fig. 2) and are listed in Table 1. All the particles are spherical and their diameters vary in the range 2 - 10 nm.

Table- I: Characteristics of CFNPs

Sample	C (% wt.)	d_{avg} (nm)	Ms (emu/g)	Hc (Oe)
M1	0.5	5.7	22.5	0
M2	1.0			
M3	5.0			
M4	7.0			

The room temperature magnetizations of the synthesized nanoparticles are shown in Fig. 3. The saturation magnetization (Ms) and coercivity (Hc) are reported in Table I. It can be seen that the prepared particles show a typical behavior of superparamagnetic particles with no coercivity and saturation magnetization of 22.5 emu/g at 15 kOe.

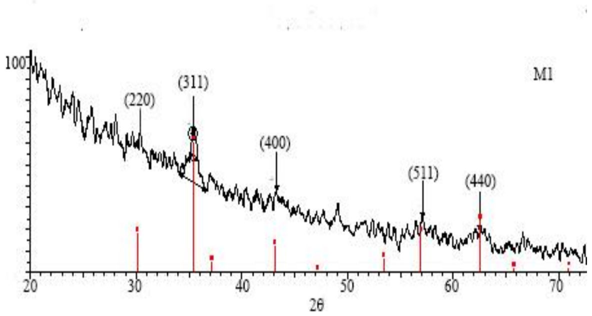

Fig. 1.XRD patterns of the synthesized particles

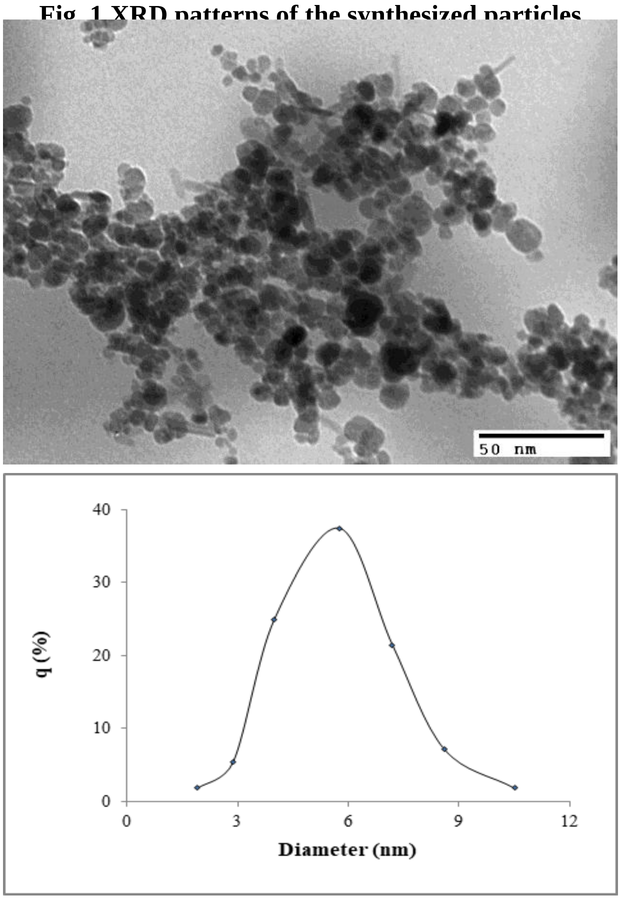

Fig. 2.TEM images and PSDs of the synthesized CFNPs

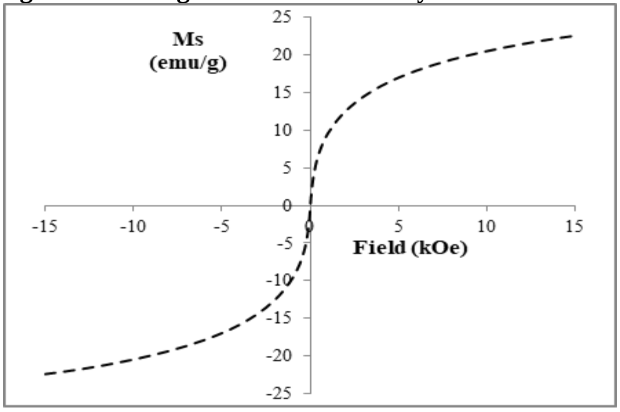

Fig. 3.Magnetization curves of CFNPs

The particle concentration, C (Table I), was calculated from TGA measurements. As seen in Fig. 4, the TGA curve of a typical fluid indicates one endothermic peak at ~ 150 °C, which may be attributed to the evaporation of water.

The residual mass is considered as the amount of cobalt ferrite particles.

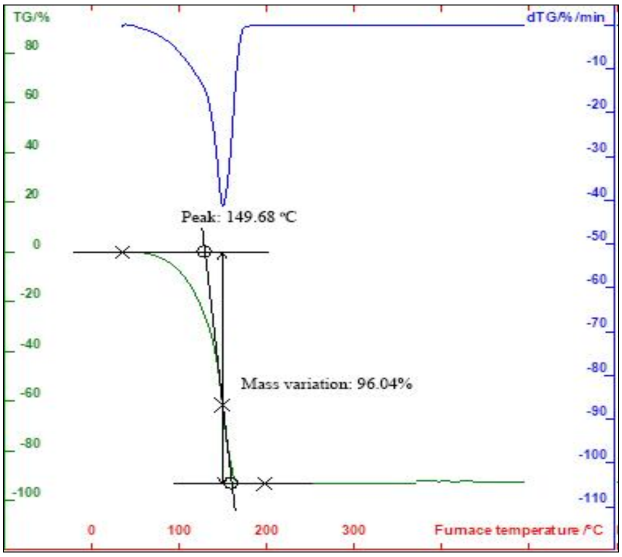

Fig. 4.Weight loss curves of a typical nanofluid (M3)

B. Temperature-dependent viscosity

A non-linear correlation between shear stress and shear rate of the fluids is presented in Fig. 5a. The result indicates that the investigated fluids are non-Newton fluids.

The viscosity of these fluids decreases with increasing shear rate (Fig. 5b), showing the shear-thinning behavior. This behavior is observed in the fluids at all the temperatures without the external magnetic field.

Fig. 5 also shows the shear stress and the viscosity decrease as the temperature is increased. The reduction in viscosity depends on the shear rate.

For example, with increasing temperature from 25 to 50 ^{o}C, the viscosity decreases from 9.17 to 5.11 mPa.s (4.06 mPa. s) at 2.8 s^{-1} and from 2.04 to 0.97 mPa.s (1.07 mPa. s) at 21.5 s^{-1}.

The increase of viscosity is attributed to the reduction of the molecular interactions between the nanoparticles and their base fluids when the temperature is increased [10].

On the other hand, with the increasing temperature, the Brownian motion of the particles is strengthened, which increases the interaction between solid particles and, thus, enhances viscosity [7].

In this study, the obtained results indicate that the first effect is more important, resulting in a decrease of viscosity. Malekzadeh, in the research for Fe_3O_4 ferrofluids [7], supposed that the decrease of the carrier liquid viscosity with an increase of temperature determines the viscosity reduction of the fluids.

This explanation is not reasonable for our results by the fact that the obtained decrease of 4.06 mPa.s is much larger than the viscosity of water. At the lower shear rates, the disordered arrangement of CFNPs is not yet broken under shear stress, the gradual decrease of viscosity is dominated by temperature. When the shear rate is enhanced, CFNPs arrange along the flow direction, and from that, only the effect of shear rate is active.

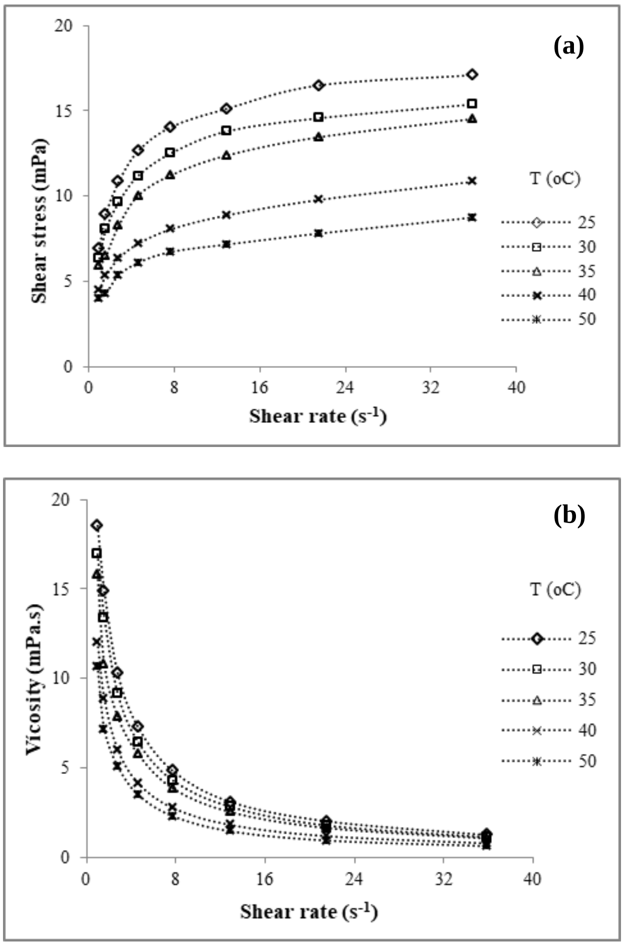

Fig. 5.Shear stress (τ) and viscosity (η) plotted vs. shear rate dependence at different temperatures (M3)

The effect of temperature on viscosity under no magnetic field is presented in Fig. 6 for various $CoFe_2O_4$ weight fractions. The viscosities were measured at the shear rate of 12.9 s^{-1}. The experimental results imply that the viscosity of the ferrofluids decreases exponentially with the temperature. The increase of viscosity is found as the nanoparticle weight fraction is increased to all the temperatures under study. However, the enhancement is not noticeable. For example, the viscosity increases from 1.33 mPa.s to 1.66 mPa.s with the range of concentration from 0.5% to 7% weight fraction at 50 °C temperature. In general, the viscosity increases as the volume concentration of particles is increased due to the rise of interaction between solid particles. However, as mentioned above, the effect of particle interactions is less important than the intermolecular effect of the solid/liquid interface. Therefore, the effect of concentration is weaker than temperature. Similar results were reported in the literature for iron oxide/ITO100 fluids [5] and $CoFe_2O_4$/EG ferrofluids [8].

The viscosity-temperature dependence was investigated at different magnetic field intensities (Fig. 7). It can be learned that the viscosity dependence on temperature is not affected significantly by the magnetic field. The field impact is more apparent at the higher particle concentrations (Fig. 7a compared Fig. 7b). As known that, with the presence of a magnetic field, the particles aggregate to form chain-like or drop-like structures along the direction of the magnetic field [11].

This arrangement becomes more potent when the magnetic field increases and the particle concentration is higher. Thus, the viscosity is enhanced. The results confirm that the effect of temperature on viscosity is substantial so that the viscosity-temperature dependence is not much influenced by other factors such as particle concentration and magnetic field [7].

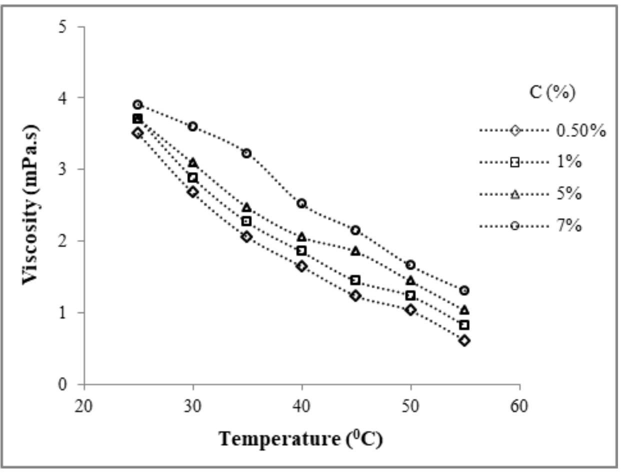

Fig. 6.Viscosity (η) shown with increasing temperature at different particle concentrations

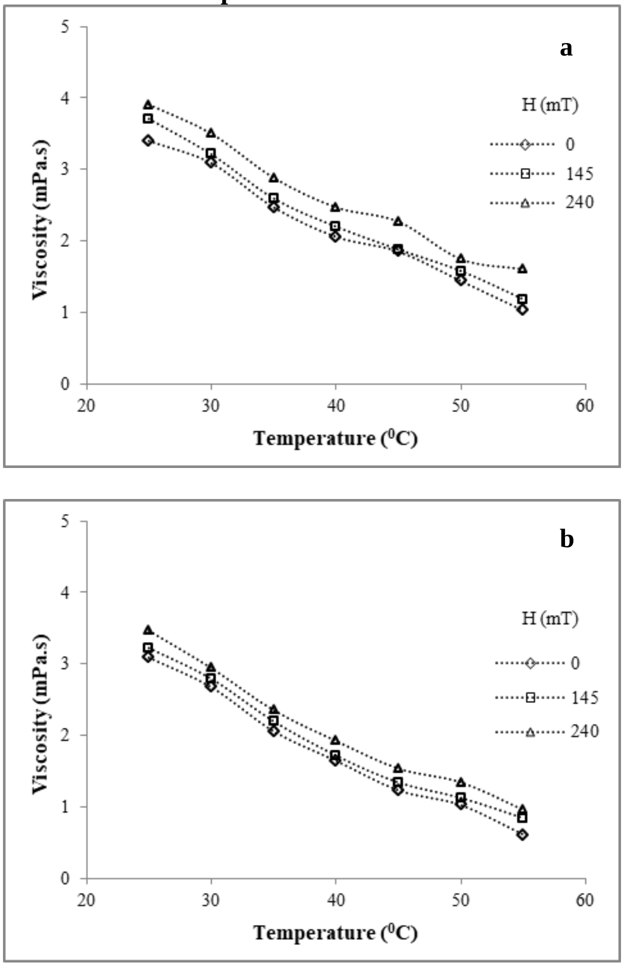

Fig. 7.Viscosity (η) shown with increasing temperature at the external magnetic field of various magnitudes. (a) Sample M3 ; (b) Sample M1

C. Constitutive equation of viscosity-temperature dependence

As seen in Fig. 8, the correlation between the logarithm of viscosity, ln(η), and the reciprocal of absolute temperature, 1/T, is linear. Therefore, the experimental data of the viscosity-temperature relation could be fitted with the Arrhenius type equation [12]

$$\ln(\eta) = \ln(A_s) + \frac{E_a}{R}\left(\frac{1}{T}\right) \tag{1}$$

Where R = 8.314 J/mol·K is the gas constant, Ea is the flow activation energy, and As is the preexponential factor of Arrhenius equation

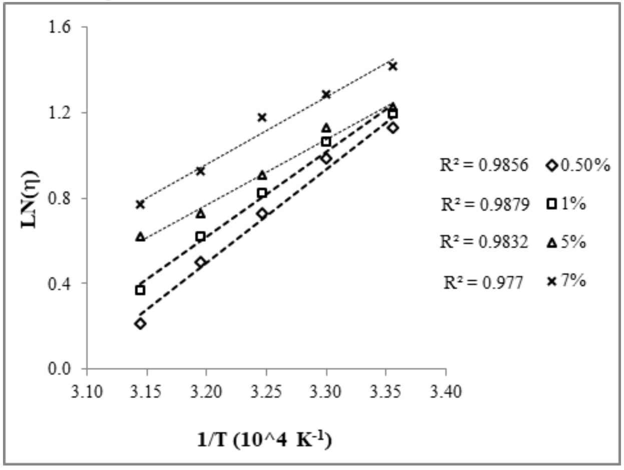

Fig. 8.Logarithm of viscosity vs. reciprocal temperature

From the plot of the logarithm of viscosity versus the reciprocal of absolute temperatures ranged from 298 to 328 K, using the linear least-squares fitting method, two parameters of the equation (1) are estimated as follows. The Ea/R is the slope of the straight line fitting to the plot. The ln(As) is the internship of the fitting line with the ordinate. Besides, the Arrhenius temperature parameter is deduced from the following formula [12]

$$T_A = \frac{-E_a}{R\ln(A_s)} \tag{2}$$

The calculated values of E_a, A_s, and T_A corresponding to the cobalt ferrite fluids with various concentrations are summarized in Table II.

Table- II: Characteristics of investigated samples

Sample	C (% wt.)	Ln(A_s)	E_a (kJ/mol)	T_A (K)
M1	0.5	-13.53	36.40	324
M2	1.0	-12.07	32.91	328
M3	5.0	-9.04	25.48	339
M4	7.0	-9.04	25.97	346

IV. CONCLUSION

Cobalt ferrite nanoparticles were successfully prepared using the coprecipitation method. The XRD and TEM analysis confirmed that the synthesized magnetic nanoparticles have a cubic spinel structure of Cobalt ferrite with an average diameter of 5.7 nm. The particles show superparamagnetic behavior with the coercivity Hc = 0 and the saturation magnetization of 22.5 emu/g. The effect of temperature on the viscosity was investigated for the CFFs with different concentrations of solid particles.

The investigations were carried out under the magnetic with various magnitudes. All the fluids show shear thinning behavior. The viscosity decreases exponentially when the temperature is increased. The viscosity-temperature dependence is not influenced significantly by either particle concentration and magnetic field. Based on the experimental results, the Arrhenius equation for the viscosity-temperature relationship is constitutive.

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