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Heat Transfer of Ferrofluids

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Abstract

Magnetic field-responsive materials are an important group of smart materials. They can adaptively change their physical properties due to external magnetic field. Magnetic liquids or ferrofluids are colloidal systems of ferro or ferrimagnetic single domain nanoparticles that are dispersed either in aqueous or in organic liquids. Currently, the research field is undergoing a transition taking into account the bulk forces in fluids, which are magnetically nonuniform. These researches enable scientists to develop promising new designs. Today, because of the advancement in technology and limited energy sources, engineering innovations are focused on development of alternative resources instead of current systems. In this chapter, it is aimed to give a brief review of the heat transfer of magnetic fluids based on different types of magnetic nanoparticles as well as some of the research and results of the heat transfer of magnetite-based ferrofluids. The heat transfer of these materials was investigated under stationary conditions, and the heat transfer coefficient was calculated.

Keywords: ferrofluid, thermal conductivity, magnetic nanoparticles

1. Introduction

Traditional heat transfer fluids such as water, oil, and ethylene glycol cause problems in the performance of engineering equipment such as heat exchangers and electronic devices due to their low thermal conductivity. To improve the performance of these devices, fluids with higher thermal conductivity have to substitute these fluids. An inventive way to increase the thermal conductivity of these fluids can be achieved by the use of the nanofluids [1]. A nanofluid is a new class of heat transfer fluids containing nanoparticles with the size range under 100 nm that are uniformly and stably suspended in a liquid. Compared to the thermal conductivity of the base fluids, nanofluids showed dramatic increase in the heat transfer due to the higher thermal



conductivity of these nanoparticles [2]. Intensive investigations on nanofluid containing metallic or nonmetallic nanoparticles such as TiO_2 , Al_2O_3 , Cu, CuO, Ag, and carbon nanotubes are being conducted to enhance their potential applications in heat transfer [3, 4].

Among different kinds of researches on nanofluids containing metallic or nonmetallic nanoparticles, some of the studies have been focused on the nanofluids prepared by dispersing magnetic nanoparticles in a carrier liquid. These are called *ferrofluids*. They are colloidal suspensions of ultrafine single domain superparamagnetic nanoparticles of metallic materials (ferromagnetic materials) such as iron, cobalt, and nickel as well as their oxides (ferrimagnetic materials) such as magnetite (Fe₃O₄) and ferrites (MnZn, Co ferrites) in either polar or nonpolar liquid carriers [5–7]. These magnetic fluids are a specific subset of smart materials that can adaptively change their physical properties under an externally applied magnetic field [6].

The research and development on the preparation, characterization, and application of the ferrofluids have been studied since mid-1960s which involve multidisciplinary sciences of chemistry, fluid mechanics, and magnetism. The most important advantage of these fluids is their ability to achieve a wide range of viscosity in a fraction of millisecond. The viscosity in the absence of magnetic field is called the "off-state" viscosity. The off-state viscosity of ferrofluids can go up to 2–500 mPa s depending on the concentration of the solid particles and the carrier liquid. Although they can respond to the action of external magnetic fields, stable ferrofluids show a relatively modest magneto-rheological effect such as an increase in yield strength. Since the particle size of the magnetic phase is very small, under ordinary field strengths, thermal agitation gives rise to Brownian forces that can overcome the alignment of the dipoles. Therefore, ferrofluids exhibit field dependent viscosity, but they exhibit no yield stress (τ_y = 0) under magnetic field. Some properties of the ferrofluid are given in **Table 1**. The field dependent viscosity is given by Eq. 1 [8].

$$\frac{\Delta\eta}{\eta} = \frac{3}{2}\varphi \frac{\frac{1}{2}\alpha L(\alpha)}{1 + \frac{1}{2}\alpha L(\alpha)} sin^2\beta \tag{1}$$

where $\alpha = \mu_0 M_d HV/kT$.

Magnetic control of the properties and behavior of these fluids are promising fields for advanced applications and a challenge for basic research and what makes these materials interesting. They are widely used in dynamic loudspeakers, computer hardware, dynamic sealing, electronic packaging, aerospace, and bioengineering [9]. Another important technological application of magnetic fluids, which depends on the heat transfer, is its use as a voice coil coolant for modern loudspeakers, high power electric transformers, and in advanced energy conversion systems like solar collectors and magnetically controlled thermosyphons [10–12]. In some of the home appliances, such as refrigerators and ovens, heat transfer techniques are used in order to provide heating or cooling. Controlling the heat transfer in these appliances with these fluids may decrease the energy consumption.

| | Ferrofluid |
|-----------------------------|--|
| Particle material | Ceramics, ferrites, iron, cobalt, etc. |
| Particle size | 5–10 nm |
| Suspending fluid | Oils, water |
| Density (gr/cm³) | 1–2 |
| Off state viscosity (mPa.s) | 2–500 |
| Required field | ~1 kOe |
| Field induced fluids | $\Delta\eta$ (B)/ η (0)~2 |
| Device excitation | Permanent magnet |

Table 1. Some of the properties of ferrofluids [6].

In the conventional nanofluids, the origin of the enhancement of thermal conductivity was thought to be due to the higher thermal conductivity of the nanoparticles (TiO₂, Al₂O₃, Cu, etc.) than the carrier fluid. Since the thermal conductivity of common magnetic materials (Fe₃O₄) is relatively low, the investigations did not gain much attention until it was understood that the thermal conductivity of the solid material did not have much effect in the enhancement of the thermal conductivity of the dispersion [13]. By understanding the control of the thermal conductivity of the ferrofluid by magnetic field increased the intensity of the research. Using ferrofluids under an applied magnetic field for the heat transfer enhancement is more advantageous compared with the conventional nanofluids (nonmagnetic nanofluids). The advantages of the ferrofluids over conventional nanofluids can be summarized as thermomagnetic convection is more intense than the gravitational one, and the thermal conductivity and viscosity are tunable under magnetic field.

2. Preparation of ferrofluids

Although pure metals (Fe, Co, Ni) possess the highest saturation magnetization, they are extremely sensitive to oxidation, hence the magnetic particles such as ferrites like magnetite (Fe_3O_4) , maghemite $(\gamma - Fe_2O_3)$, or others (stoichiometric formula: $MO \cdot Fe_2O_3$, where M is a divalent ion, M = Mn, Zn, Ni, Co, Fe) are commonly used in ferrofluids. And among these, nano-sized iron oxide is the most widely used magnetic phase in ferrofluids. Various approaches have been explored for synthesis and characterization of high quality magnetic iron oxide nanoparticles. For example, sol-gel pyrolysis method was performed by Laokul et al. [14]. Synthesis of nanoparticles by thermal reductive decomposition method was performed by various scientists [15, 16]. Waje et al. performed mechanical alloying technique [17]. Hydrothermal technique was also used by various scientists to synthesize ferrite nanoparticles [18, 19]. However, the chemical method of coprecipitation of ferrous and ferric ions from solutions by addition of an alkali is a method which is very often used to prepare nanoparticles due to its low cost and simplicity [20]. Size reduction could be another method where magnetic powder of micron size is mixed with a solvent and a dispersant in a ball mill in order to grind for a period of several weeks [21].

The function of the carrier liquid is to provide a medium in which the magnetic powder is suspended. Ferrofluids used in different research and technology fields have been synthesized in carrier liquids such as water, silicone oil, synthetic or semi-synthetic oil, mineral oil, lubricating oil, kerosene, and combinations of these and many other polar liquids [22–24]. Boiling temperature, vapor pressure at elevated temperature, and freezing point are important parameters to be considered when choosing the carrier liquid. The carrier liquid should be non-reactive with the magnetic phase and also with the material used in the device. In terms of the heat transfer applications, the choice of the carrier fluid for the ferrofluid needs some additional requirements such as high conductivity, high heat capacity, and high thermal expansion coefficient. Water, oils, and ethylene glycol are considered as conventional heat transfer fluids and these can be good candidates for the carrier liquids. In recent years, studies on ferrofluids using ionic liquids have been reported, which seems to be a promising field of study [23, 24].

Colloidal stability of the ferrofluids is important in the technological applications. The stability is obtained by minimizing the agglomeration, which is maintained by the addition of the surfactants. The additives must be chosen to match the dielectric properties of the carrier liquid. Various surfactants such as silica, chitosan, polyvinyl alcohol (PVA), and ethylene glycol are usually used to coat the nanoparticles and to enhance dispersibility in aqueous medium [25–28]. Oleic acid (OA) is a commonly used surfactant to stabilize magnetic nanoparticles synthesized by traditional coprecipitation method [22]. Antioxidation additives may also be added to prevent oxidation. In water-based MR fluids, pH control additives are also used.

Magnetic nanoparticles tend to aggregate due to strong magnetic dipole-dipole attraction between particles. Stability of the magnetic colloid depends on the thermal contribution and the balance between attractive (van der Waals and dipole-dipole) and repulsive (steric and electrostatic) interactions. Under the magnetic field, the magnetic energy derives the particles to higher intensity regions; on the other hand, thermal energy forces the particles to wander around in the whole liquid. The stability against segregation is favored by the high ratio of the thermal energy to the magnetic energy. Stability against settling due to gravitational field is given by the ratio between gravitational energy and magnetic energy [8]. The two basic attractive interactions between the magnetic particles are dipole-dipole and van der Waals-London interactions. The ratio of thermal energy (kT) to dipole-dipole contact energy ($E_{\text{dipole}} = (\mu_0 M^2/12)V$) must be greater than unity. The particle diameter is given by $D \le (72 kT/\pi \mu_0 M^2)^{1/3}$, and the particle size is calculated as $D \le 7.8$ nm. The normal ferrofluids with the particle size of 10 nm are in the limits of agglomeration. Van der Waals forces arise due to the fluctuating electric dipole-dipole forces. Preventing the contact of the particles is another necessity if a stable colloid is to be obtained [8].

The Brownian motion, electrostatic repulsion, and steric repulsion are the main mechanisms supporting the ferrofluid colloidal stability. Electrostatic interaction is the dominant mechanism in ionic ferrofluids, whereas steric repulsion is the dominant mechanism supporting the colloidal stability in organic-based ferrofluids [23]. The agglomeration of particles suspended in a liquid can be prevented by creating mutually repelling charged double layers or by physically preventing the close approach of particles by steric hindrance which is provided by

the surfactant molecules adsorbed onto the particle surface [8, 23]. As the thickness of the adsorbed polymer is increased, the stability of the dispersion increases [8, 23]. Wang and Huang showed that by retaining excess oleic acid in their ferrofluid, stable magnetic colloid was achieved by steric repulsion [8, 23].

3. Thermal conductivity of ferrofluids

3.1. Experimental investigations

Thermal conductivity of ferrofluids has gained much attention in the last decade due to the significant enhancement compared to the nonmagnetic nanofluids. The increase in the thermal conductivity can occur both with and without the applied magnetic field. Experimental studies show that the change in the off-state (when there is no magnetic field) thermal conductivity of ferrofluids could be due to volume fraction of magnetic phase, particle size distribution, temperature, surfactant, etc. On the other hand, when the magnetic field is applied, besides the factors mentioned above, the magnitude and direction of the applied magnetic field affect the thermal conductivity of the ferrofluids.

In the experimental studies of the thermal conductivity of ferrofluids, it has been observed that both the on-state and off-state thermal conductivities increase with the increase in the volume fraction of the magnetic phase. When the literature was reviewed, it was seen that most of the ferrofluids synthesized with magnetite (Fe₃O₄) which was produced by copreciptation method has been studied. Abareshi and coworkers synthesized ferrofluids by dispersing Fe₃O₄ nanoparticles in water [29]. They reported an increase of 11.5% in the offstate thermal conductivity as the particle loading increased to 3 vol% at 40°C. This increase was observed when the magnetic field was applied parallel to the heat flux. The study of Li et al. was also performed with water-based ferrofluids and an increase in the on-state (magnetic field, H = 19 kA/m) thermal conductivity of 11% for 1 vol% and 25% for 5 vol% magnetic nanoparticles was reported [30]. Philip et al. and Shima et al. [30, 31] investigated the thermal conductivity of kerosene-based ferrofluids synthesized with Fe₃O₄ nanoparticles. When the magnetic field was applied parallel to the heat flux, they discovered a dramatic increase in the thermal conductivity. For a volume fraction of 6.3%, the increase was 300% at H = 7 kA/m field strength. No increase was observed when the magnetic field was applied perpendicular to the heat flux. The reason may be that the different directions of external magnetic field lead to quite different morphologies of the magnetic fluids that exerted quite different effects on the energy transport process inside the magnetic fluid [31]. They further explained that the chains formed by the particles provided more effective bridges for energy transport inside the ferrofluid along the direction of temperature gradient and as a result, the thermal process in the ferrofluid was enhanced. The anisotropic property of thermal conductivity was addressed in the theoretical study by Fu et al., and Blums et al [10, 32]. Blums et al. predict anisotropy of thermal conductivity in ferrofluids in the presence of a magnetic field [10]. In the research conducted by Gavili et al. the thermal conductivity of ferrofluids containing Fe₃O₄ nanoparticles suspended in deionized water under magnetic field was experimentally investigated [33]. According to their results, a ferrofluid with 5.0% volume fraction of nanoparticles with an average diameter of 10 nm enhanced the thermal conductivity more than 200% at 1000 Gauss magnetic field [33].

As it is seen from all the research and reports, it is evident the experimental results have been heterogeneous. The difference in the experimental outcomes may be due to magnetization, size distribution of the particles, the type of the carrier liquid, etc. The effect of the carrier liquid showed that the thermal conductivity ratio is higher for carrier liquid with a low thermal conductivity like common hydrocarbons. However, the absolute thermal conductivity of ferrofluid is higher for a carrier liquid with a high thermal conductivity.

In recent years, studies have been carried out to understand the thermal conductivity of ferrofluids synthesized by magnetic phase other than Fe₃O₄, especially with carbon nanotubes (CNT) has also been investigated by many scientists. Hong et al. [34] and Wensel et al. [35] experimentally measured the thermal conductivity of single wall carbon nanotubes coated by Fe₂O₃ nanoparticles suspended in water and they observed an approximately 10% increase in the thermal conductivity with 0.02% particle loading. Wright et al. reported thermal conductivity enhancement of single wall carbon nanotubes coated by Ni nanoparticles suspended in water [36]. Sundar et al. measured the thermal conductivity enhancement of the hybrid ferrofluid which was composed of carbon nanotube (CNT)—Fe₃O₄ and water [37]. They observed a thermal conductivity enhancement of 13.88–28.46% at 0.3% volume concentration in the temperature range of 25–60°C. Shahsavar et al. analyzed the thermal conductivity behavior of Fe₃O₄ and CNT hybrid ferrofluids and observed that the highest enhancement in the thermal conductivity was about 151% for 0.9% ferrofluid and 1.35% CNT [38].

The review of the literature on the experimental studies of the thermal conductivity of ferrofluids revealed that the thermal conductivity is enhanced by the volume fraction of the magnetic phase and the applied magnetic field. Next chapter will discuss the reasons for the abnormal enhancement in the thermal conductivity of the ferrofluid under the influence of applied magnetic field.

3.2. Mechanisms of heat transfer enhancement

In the thermal conductivity of conventional nanofluids and ferrofluids, the most discussed mechanisms have been Brownian motion and formation of particle chain/cluster structure [39]. The Brownian motion indicates the random movement of particles dispersed in liquid or gas, and the motion is due to collision with base fluid molecules, which makes particles undergo a random walk motion [40]. The Brownian motion could contribute to the thermal conductivity enhancement in two ways, namely, the direct contribution due to motion of nanoparticles that transports heat (diffusion of nanoparticles) and the indirect contribution due to the so called micro-convection of fluid surrounding individual nanoparticles [40]. The diffusion of magnetic nanoparticles plays an important role at a low volume fraction (φ < 2%), which could be explained by the effective medium (Maxwell) theory rather than the effects associated with the Brownian motion-induced hydrodynamics. The effective medium or mean-field theory of Maxwell, which describes the effective macroscopic properties of the composite material as a function of the particle fraction and the material properties of the components, is most often

used to analyze the thermal conductivity results of nanofluid experiments. For a nanofluid with non-interacting spherical nanoparticles with low volume fraction, the theory predicts (Eq. 2)

$$\frac{\kappa}{\kappa_f} = \frac{1 + 2\beta\Phi}{1 - \beta\Phi} \tag{2}$$

where φ is the nanoparticle volume fraction, κ_p and κ_f are thermal conductivity of the particle and the fluid, respectively. $\beta = (\kappa_p - \kappa_f)/(\kappa_p + 2\kappa_f)$, and $(\kappa_p - \kappa_f)$ is the difference between the thermal conductivities of the nanoparticle and the base fluid. However, in the study done by Vadasz et al. [41] and Keblinski et al. [42], the results of the thermal conductivity measurements showed divergence from the effective medium theory. One possible discrepancy between effective medium theory and the experimental results is the interparticle interactions, which can result in the formation of chain and cluster-like formations. Philip and coworkers showed that the micro-convection of the fluid medium around randomly moving nanoparticles did not affect the thermal conductivity of a nanofluid and the microconvection model overestimated the thermal conductivity values [43]. According to them, the conductivity enhancement in the ferrofluid at high volume fraction (φ < 2%) was due to the presence of dimmers or trimmers in the fluid. These results were in a reasonable agreement with the Maxwell-Gannet model, especially at higher volume fractions. Clusters or chains of the particles may form heat bridges [31]. The form and magnitude of these structures vary and depend not only on the material of the carrier medium and the particles but also on the shape and size of the particles [31].

In ferrofluids, the interparticle interactions have even more important impact on the properties of the fluid, due to the chain-like formation of the particles caused by the magnetic dipole interactions. The effect of this interaction can be seen, for example, in changes in the viscosity of ferrofluids, which depends on interparticle interaction [7, 44].

Magnetically induced structure formation only arises if the magnetic energy of the particles is larger than their thermal energy. The mechanism of thermal conductivity enhancement can be explained as follows: The magnetic particles in the ferrofluid are single domain and superparamagnetic with magnetic moment m as mentioned above [28]. The interparticle dipole-dipole interaction, which is also called dipolar coupling, refers to the interaction between magnetic dipoles. The potential energy of the interaction U_d is given by Eq. (3),

$$U_d(ij) = \begin{bmatrix} 3(m_i \cdot r_{ij})(m_j \cdot r_{ij}) / r_{ij} - (m_i \cdot m_j) / r_{ij} \\ r_{ij}^5 - r_{ij} \end{bmatrix}$$
(3)

Suppose m_i and m_j are two magnetic moments in space and r_{ij} (= r_i - r_j) is the distance between the ith and the jth particles. The magnetic moments are oriented in random directions in the absence of magnetic field and the nanoparticles are influenced by the Brownian motion as the

thermal energy exceeds the magnetic dipole attraction ($U_d(ij) < k_BT$). In the presence of a magnetic field, the magnetic dipolar interaction becomes strong enough to dominate the thermal energy so that the magnetic particles start aligning in the direction of the magnetic field [31]. **Figure 1** gives the schematic drawing of the clustering/chain-like formation under magnetic field.

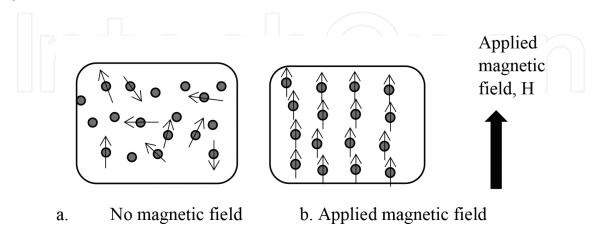


Figure 1. Schematic drawing of the chain-like formation of the magnetic particles in the fluid. (a) No magnetic field. (b) Applied magnetic field.

The lengths of the chains depend on the magnitude of the magnetic field. As the magnetic field increases, the particles start forming short chains along the direction of the magnetic field and the chains get longer as the magnetic field increases. Based on Philip's theory due to linear chain-like structures of the magnetic nanoparticles, the percolation theory could support the abnormal enhancement of the ferrofluid. They stated that the maximum enhancement was observed when the chain-like aggregates were well dispersed without clumping [31].

Although the thermal conductivity of ferrofluids enhances with increasing magnetic field, there are reports regarding a decrease in the thermal conductivity of these fluids. Shima et al. observed decrease in the thermal conductivity above 82 Gauss magnetic field [31]. They attributed this decrease to the "zippering" of the chains. The linear and thick aggregates with the aspect ratio due to zippering can collapse to the bottom of the cell, and hence the thermal conductivity cannot be measured. Gavili and coworkers observed that the thermal conductivity dramatically decreased in the presence of magnetic field with increasing temperature. When the temperature of the ferrofluid increases, the chain-like structure is broken due to the increase in the thermal velocity and consequently the thermal conductivity decreases [33].

Theoretical and experimental studies related with the thermal radiation and convection of the heat transfer of nanofluids have started to gain more attention in the recent years especially, in the field of engineering applications such as solar collectors and in space applications [45–47]. Thermal convection in magnetic fluids heated from below subjected to an external magnetic field causes a convection-driving mechanism. The temperature difference causes a gradient in the magnetic field and as a result a magnetic force appears. Beyond a certain threshold a thermomagnetic convection is generated.

4. Experimental study

4.1. Apparatus and data analysis

An experimental setup is built according to the ISO 8301 numbered "Thermal insulationdetermination of steady state thermal resistance and related properties-Heat flow meter apparatus" standard has been used in the heat transfer experiments. Experimental setup has been established as single-specimen asymmetrical configuration according to the standard shown in Figure 2.

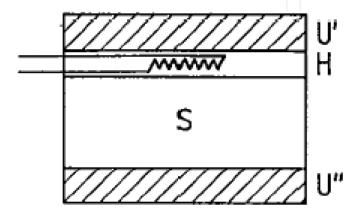


Figure 2. Single-specimen asymmetrical configuration (U', U" are the cooling and heating units, respectively, and $H = \frac{1}{2} \frac{1}{2$ heat flux meter).

Schematic drawing containing the requirements of experimental setup such as testing unit, two water baths, data acquisition system, and computer is given in Figure 3. The testing unit was heated and cooled by water bath.

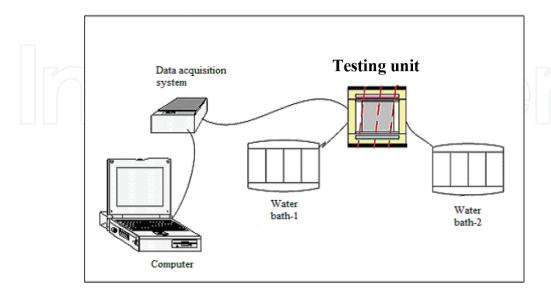


Figure 3. The illustration of experimental setup.

A more detailed representation of the testing unit given in **Figure 4**, which consisted of a space for sample, heat flux sensors, thermocouples, heating-cooling sources, and polyurethane insulating material from outside to inside. The magnetic field was applied by neodymium permanent magnets parallel to the temperature gradient. The maximum magnetic field obtained was approximately 140 Gauss. One of the uncertainties of the setup could be the non-uniformity of magnetic field. The magnetic field was calculated as the average of the field from three different points on the radial direction. Another uncertainty could be heat loss. Although the experimental setup was isolated, there could still be some heat loss. For reproducibility of the data, each measurement was performed for five ferrofluid samples.

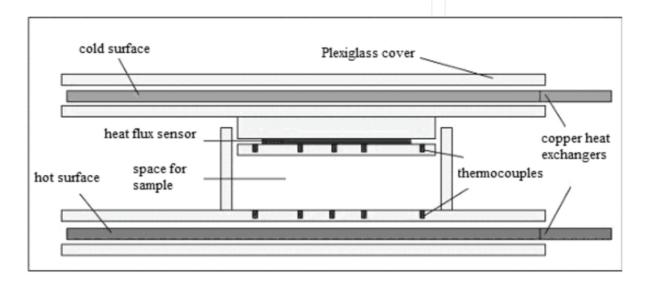


Figure 4. Testing unit.

In order to determine the temperature range at which the thermal conductivity of ferrofluids is more effective for different applications, the experiments were done in two different temperature intervals; from -20 to 0°C, and from 0 to 50°C and two different temperature differences. The temperatures difference between the hot and cold surfaces of the setup are given in **Table 2**.

| Between 0 and -20°C interval | Between 0 and 50°C interval |
|------------------------------|------------------------------|
| Temperature difference, 20 K | Temperature difference, 35 K |
| Temperature difference, 15 K | Temperature difference, 20 K |

Table 2. Temperature intervals and temperature differences at which the experiments were performed.

Besides the effect of temperature, following variables were considered during investigation of heat transfer of ferrofluids, different carrier liquid and concentration of the magnetic phase.

The thermal conductivity measurements have been carried out when the water baths came a steady state temperature. The heat transfer coefficient has been calculated by using Eq. 4,

$$\kappa = \frac{A}{Q} \frac{\Delta T}{\Delta x} \tag{4}$$

where *A* is the cross-sectional area, *Q* is heat flux, ΔT is the temperature difference, and Δx is the height of the measuring cell.

4.2. Ferrofluid preparation

Synthesis of stable and well dispersible MR fluid is extremely important for mechanical and heat transfer applications. Synthesis of stable ferrofluid depends on concentration and viscosity of carrier fluid, concentration of magnetic phase, particle size, and surfactants. The ferrofluids in this study were synthesized in water and silicone oil (viscosity 350 cSt) with as received magnetite Fe₃O₄ nanopowder from Aldrich. The particle size was approximately 50 nm. Samples were synthesized as volumetric percentages of 5 and 20%. Surfactant was also added to prevent sedimentation. The names and description of the ferrofluids are given in Table 3.

| Name | Description |
|--------------------------------------|--|
| 5 Fe ₃ O ₄ -S | 5 vol% Fe_3O_4 + 350 cSt silicone oil + surfactant |
| $5 \text{ Fe}_3\text{O}_4\text{-W}$ | $5 \text{ vol}\% \text{ Fe}_3\text{O}_4$ + water |
| 20 Fe ₃ O ₄ -W | $20 \text{ vol}\% \text{ Fe}_3\text{O}_4$ + water |
| 20 Fe ₃ O ₄ -S | $20 \text{ vol}\% \text{ Fe}_3\text{O}_4 + 350 \text{ cSt silicone oil} + \text{surfactant}$ |

Table 3. Description of the MR fluids used in this research.

In order to have a stable dispersion ball milling which could break up the agglomerated, was applied. Ball milling procedure was conducted with yttria stabilized zirconia grinding media with 0.5 mm diameter.

5. Results and discussion

5.1. Analysis of thermal conductivity in the temperature interval between 0 and -50°C.

In this part of the study, base liquid, volume fraction (5 and 20 vol% Fe_3O_4), temperature (ΔT = 20 and 35 K), and dependence of the thermal conductivity of the ferrofluids were investigated.

5.2. Carrier liquid dependence of the 5 vol% Fe₃O₄-based ferrofluids

Heat transfer coefficients of ferrofluids were analyzed for two different temperature intervals, and in each temperature interval, the thermal conductivity of the ferrofluids was investigated for two different temperature differences. The temperature intervals were chosen as 0 to 50°C and -20 to 0°C, and the temperature differences are ΔT = 35 K and ΔT = 20 K for the first interval and ΔT = 15 K and ΔT = 20 K for the second interval mentioned above.

In the first part of the study, the heat transfer was investigated for $5\text{Fe}_3\text{O}_4\text{-S}$ and $5\text{Fe}_3\text{O}_4\text{-W}$ at a temperature difference of 20 K. The increase in heat transfer coefficient in the presence of magnetic field of 134 Gauss was 7 and 18% for $5\text{ Fe}_3\text{O}_4\text{-S}$ and $5\text{ Fe}_3\text{O}_4\text{-W}$ -type ferrofluids, respectively (**Figure 5**).

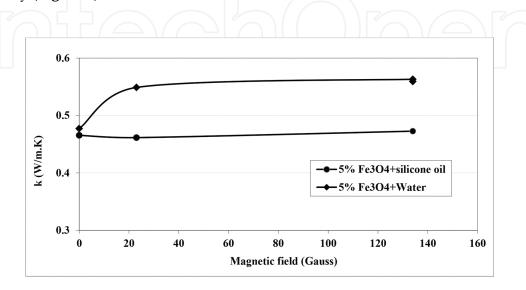


Figure 5. Thermal conductivity at 0–50°C interval and the temperature difference ΔT = 20 K.

As discussed above, the heat transfer increased as we increased the magnetic field. Although this increase could be attributed to the chain formation of the iron particles in the fluid, the effect of magnetic field is still not very clear. In **Figure 6**, the percent change in the thermal conductivity is given at temperature difference of $\Delta T = 35$ K in temperature interval of 0–50°C

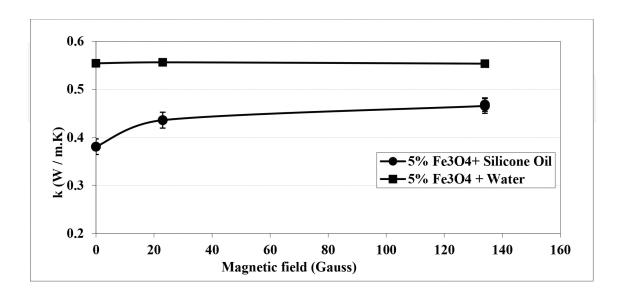


Figure 6. Thermal conductivity at 0-50°C interval and the temperature difference $\Delta T = 35$ K.

As given in **Figure 6**, heat transfer coefficients for 5 Fe₃O₄-S and 5 Fe₃O₄-W at the highest magnetic field were measured as 0.47 and 0.56 W/m K, respectively. In addition, the increase in the thermal conductivity for 5 Fe₃O₄-S was about 23% and water based was 5%. In either temperature differences (ΔT = 35 and 20 K), the thermal conductivity coefficient of silicone-based ferrofluid is less than that of the water-based ferrofluid. Since the volume concentration of the magnetic phase is small, the thermal conductivity of the base liquid could be a factor in the increase of the thermal conductivity of ferrofluid. The thermal conductivity of water is more than that of the silicone oil.

5.3. Volume percent and temperature dependence of thermal conductivity

In this part of the study, the volume dependence of the thermal conductivity was investigated. This investigation was performed at ΔT = 35 and 20 K as well. In **Figure 7**, it is observed that as the volume fraction is increased, the thermal conductivity coefficients for 5 Fe₃O₄-S and 20 Fe₃O₄-S ferrofluid also increased for ΔT = 35 K. At the highest magnetic field, the thermal conductivity coefficient, **k**, is measured as 0.51 and 0.47 W/mK for 20 and 5 vol%, respectively. The percent increase in both of the fluids was the same, which was approximately 24%.

When the temperature difference is kept smaller (ΔT = 20 K) in the same temperature interval, the thermal conductivity coefficient, **k**, is 0.51 and 0.47 W/m K (**Figure 10**) at 134 Gauss which were the same as the coefficients in ΔT = 35 K and the percent change of the thermal conductivity for these two intervals was almost the same. When we compare **Figures 7** and **8** at a magnetic field of 134 Gauss we saw that the **k** values were the same for 20 and 35 K temperature differences. However, at zero magnetic field, they were different which made a difference in the percent increase in the 35 K difference is more than that of the 20 K difference.

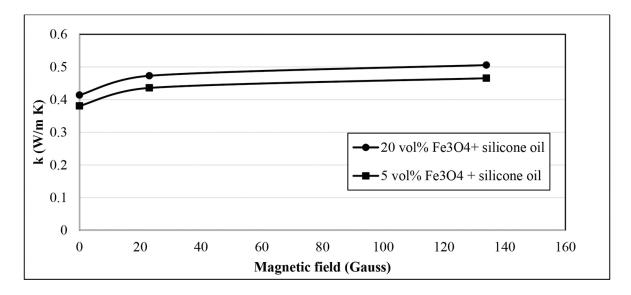


Figure 7. Volume dependence of thermal conductivity between 0 and 50°C temperature interval and 35 K temperature difference.

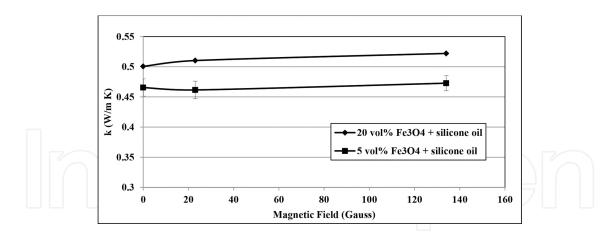


Figure 8. Volume dependence of thermal conductivity between 0 and 50°C temperature interval and 20 K temperature difference.

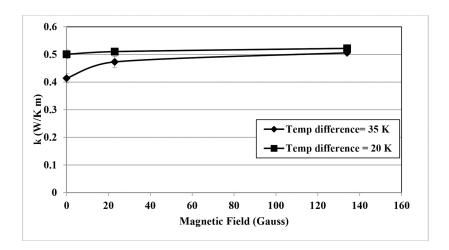


Figure 9. Temperature difference dependence of 20 vol% Fe_3O_4 and silicone oil-based ferrofluid.

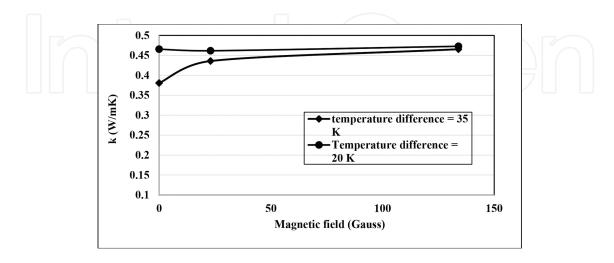


Figure 10. Temperature difference dependence of 5 vol% Fe₃O₄ and silicone oil-based ferrofluid.

Figures 9 and **10** give a clearer view of the temperature difference dependence of the thermal conductivity for 20 $\text{Fe}_3\text{O}_4\text{-S}$ and 5 $\text{Fe}_3\text{O}_4\text{-S}$ -type ferrofluid, respectively. In both graphs, the thermal conductivity enhancement is more for 35 K temperature difference. As the magnetic field increased, the k values reached the same value irrespective of the temperature difference.

5.4. Analysis of thermal conductivity in the temperature interval between -20 and 0°C.

In this part of the study, the thermal conductivity of the for $5 \, \text{Fe}_3 \text{O}_4$ -S and $20 \, \text{Fe}_3 \text{O}_4$ -S ferrofluids were investigated in the temperature interval of -20 to 0°C in which the temperature differences were kept as 15 and $20 \, \text{K}$.

5.4.1. Volume percent and temperature dependence of thermal conductivity

The next set of measurements involved a lower temperature interval, such as from -20 to 0° C. In this interval, the temperature differences were taken as 15 and 20 K. **Figure 11** shows the change in the thermal conductivity with respect to the magnetic field when the temperature gradient was 20 K. The percentage decrease in the thermal conductivity of ferrofluids for 5 and 20 vol% ferrofluids at 134 Gauss was 33 and 34%, respectively. The volume dependence of the thermal conductivity was also observed in these measurements. The 20 vol% ferrofluid had a higher thermal conductivity. **Figure 12** shows the change in the thermal conductivity with respect to the magnetic field under a 15 K temperature difference. Unlike the results obtained in the higher temperature intervals, there was a very small increase in the conductivity, followed by a slight decrease as the magnetic field increased. All the fluids showed a similar trend. The same amount of decrease was observed in this range for two different magnetic phase concentrations. The percentage decrease in the thermal conductivity for 5 Fe₃O₄-S and 20 Fe₃O₄-S type ferrofluids at 134 Gauss was 2 and 3%, respectively. The volume dependence

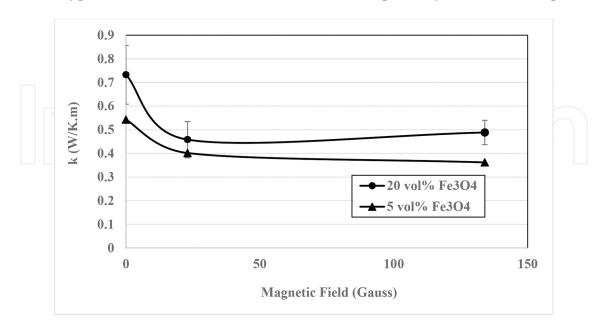


Figure 11. Volume dependence of thermal conductivity between -20 and 0°C temperature interval and 20 K temperature difference.

of the thermal conductivity was also observed in these measurements. The for 20 Fe_3O_4 -S type ferrofluid had a thermal conductivity of 42 W/K.m at 134 Gauss whereas for 5 Fe_3O_4 -S type ferrofluid had 37 W/K.m.

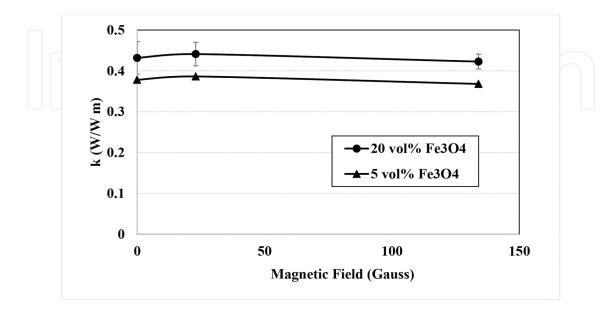


Figure 12. Volume dependence of thermal conductivity between -20 and 0°C temperature interval and temperature difference of 15 K.

The thermal conductivity depended on the temperature difference at very low magnetic fields. As the magnetic field increases the thermal conductivity became irrespective of the temperature. This behavior is observed both for 5 Fe₃O₄-S and 20 Fe₃O₄-S (**Figures 13** and **14**, respectively).

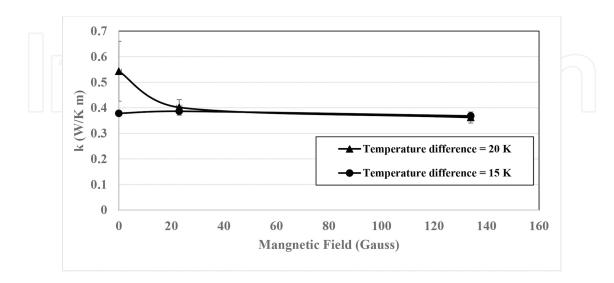


Figure 13. Temperature difference dependence of 5 vol% Fe₃O₄ and silicone oil-based ferrofluid (5Fe₃O₄-S).

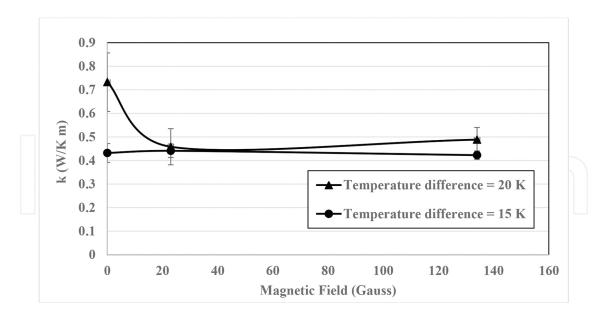


Figure 14. Temperature difference dependence of 20 vol% Fe₃O₄ and silicone oil-based ferrofluid (20Fe₃O₄-S).

6. Conclusion

Thermal conductivity mechanism in liquid involves collision of the molecules and transfer of the energy and momentum with one another. Transfer of the kinetic energy occurs in the lower temperature part of the system when a molecule moves from a high temperature region to a region of lower temperature and this molecule gives up this energy via collision with lower energy molecules. In solids, on the other hand, thermal energy may be conducted by lattice vibrations. At low-temperature ranges (-20 to 0°C), the energy of the molecules in the liquid is not enough to cause collisions and lattice vibrations could be insufficient to conduct heat. The decrease of the thermal conductivity observed in the temperature range between 0 and -20°C could be due to the less energetic particles. We consider that the thermal conductivity of the ferrofluid is determined by the factors such as stability of the ferrofluid particle size and the viscosity of the base liquid. Another important point in the heat transfer of the ferrofluids could be the thermo-convective instability of the magnetic fluids. The instability arises due to the stronger magnetization of the colder fluid which is drawn to the higher region and is displaced by the warmer fluid [5, 46, 47]

As the temperature decreases the density and viscosity of the base fluid, silicone oil, increase. The denser and more viscous fluid hinders the motion of the magnetic particles due to the temperature difference. Thus, lower temperatures inhibit the motion of particles in the ferrofluid which will prevent settling of the particles. The stability of the fluid can be affected in a negative way at higher temperatures and at lower magnetic fields because the density and viscosity of the silicone oil decrease at high temperature. The instability of the ferrofluid at low temperature could be the reason for the different thermal conductivities at low magnetic fields. At low temperature, chain formation between the magnetic particles is activated by the

increase in magnetic field. Consequently, this increases the thermal conductivity of the magnetic fluid at low temperatures. As the magnetism increases, the effect of the temperature range diminishes and thermal conductivity reaches almost the same value.

The heat transfer characteristics of $5\text{Fe}_3\text{O}_4\text{-S}$ and $20\text{Fe}_3\text{O}_4\text{-S}$ ferrofluid were investigated in the presence of the magnetic field applied parallel to the temperature gradient. The thermal conductivity behavior of the ferrofluids in different temperature ranges was analyzed, and it was seen that the heat transfer was more effective at higher temperatures. The fluids showed an increase in the thermal conductivity in the temperature intervals from 0 to 50°C, and a decrease from -20 to 0°C.

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Nomenclature

 $\Delta \eta$ (B): Change in viscosity under magnetic field (Pa s)

 η (0): Viscosity without magnetic field (Pa s)

k: Boltzmann constant (J/K)

T: Temperature (K)

 E_{dipole} : Dipole-dipole energy

 μ_0 : Vacuum permeability (Vs/A m)

M: Magnetization (A/m)

 M_d : Domain magnetization

H: Magnetic field (Gauss)

V: Volume (m³)

D: Particle diameter (m)

 κ_p : Thermal conductivity of the particle (W/m K)

 $\kappa_{\rm f}$: Thermal conductivity of the fluid

 β : Magnetic induction (T)

 Φ : Volume fraction

 $U_{\rm d}$: Potential energy of the interaction

 $m_{i,} m_{j}$: Magnetic moments in space

 r_{ij} : (= r_i - r_j) distance between the ith and the jth particle

A: Cross-sectional area

Q: Heat flux

 ΔT : Temperature gradient

 Δx : Distance between the plates (m)

 $L(\alpha)$: Langevin function

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