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Thermophysical Properties of Nanoferrofluid (Fe_3O_4 –Acetone/Znbr$_2$) as a Working Fluid for Use in Absorption Refrigeration Applications

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Abstract

In this paper, thermophysical analyses were achieved for the mixture of (Fe_3O_4-acetone/$ZnBr_2$) refrigeration working solution and examine its efficiency characteristics as a nanoferrofluid for use in absorption refrigeration applications driven by different low temperatures sources. Where it shows an investigation of the chosen nanoferrofluid containing the preparation, stability, structure, and properties. The reasons behind choosing Fe_3O_4 nanoparticles are that acetone is a good dispersivity medium for this kind of nanoparticles, also, their excellent thermophysical properties and magnetic property which give an ability to utilize them combined with applying an external magnetic field as a method for long and acceptable suspension stability of these nanoparticles in the base fluid thus enhancement in the heat transfer process in the generator of Absorption Refrigeration System (ARS). As a multi-factor experimental study, the experiments are designed to visually inspect the suspension nanoparticle's stability in the base fluid. Then presenting the thermophysical analysis of different properties of the mixture (thermal conductivity, density, dynamic viscosity, and specific heat capacity). The results elucidate that the studied nanoferrofluid has good dispersion and an enhancement in thermal conductivity that reaches 10.179 % at 0.2 (wt.%) of nanoparticle concentration. Also, by increasing nanoparticle concentration, the density increased, heat capacity decreased, as expected, and viscosity significantly increased.

Keywords: *Acetone/$ZnBr_2$; Fe_3O_4; nanoferrofluid; thermophysical properties; thermal conductivity; absorption refrigeration system.*

1. Introduction

Newly, Refrigeration binary fluids that have dispersed nanoparticles have acquired an interest in refrigeration and air conditioning fields due to their excellent thermophysical properties. These advantages make them used in these kinds of systems with many studies for performance improvement [1-5]. Yang et al. [6] prepared various types of nanofluids by mix up poly acrylic acid (PAA) and Al_2O_3 nanoparticles with poly ethylene glycol (PEG 1000), also, TiN, and SiC, with PEG 10000 to the hydroxyapatite NH_3/H_2O solution, respectively. Results showed that thermal conductivity is enhanced by increasing the nanoparticle content and the temperature and by decreasing the nanoparticle volume. Nourafkan et al. [7] have investigated the photo-thermal conversion features of a long-term and the stability against the agglomeration and sedimentation of a stable nanofluid (nanosize particles in 50 wt.% lithium bromide of water-lithium bromide base fluid). Experimental results represented that the utilize of nanofluid could considerably enhance the light-trapping efficiency and, therefore, the bulk temperature.

A unique type of nanofluids is nanoferrofluids. Nanoferrofluids content ferromagnetic nanoparticles that exhibit higher thermophysical properties, Nourafkana et al. [8]. The problem with magnetic particles is that the long-range magnetic forces always will overcome any charge ('electrostatic') stabilization attempts. Moreover, Van Der Waals' forces are always attractive. If it is wanted to prevent agglomeration, we have to modify the surface of magnetite nanoparticles. Unless it is attached to any ligand to the particles, they will tend to agglomerate eventually. One route to stabilization is steric and a polymer such as PEG or poly ethylene imine (PEI) on the nanoparticles [9]. Farhanian et al. [10] had studied a new methodology that drives to an active covering with a thickness of 1.4–10 nm, assured by High-resolution transmission electron microscopy (HRTEM) and thermogravimetry analysis (TGA). X-ray photoelectron spectroscopy (XPS) and time-of-flight secondary ion mass spectrometry (ToF-SIMS) description confirms that the covering has consisted of both aliphatic and polymerized carbon chains, with combined organometallic bands and oxygen-containing moieties. There are other studied routes to stabilization by applying magnetic vortex or sonication. The magnetic vortex method to stabilization had investigated by Wu et al. [11], the addition of a Fe_3O_4 nanoferrofluid in incorporation with the applying of an external magnetic field as a novel method for improving the absorption in an ammonia-water bubble pump absorber. Then they carried out an exhaustive experimental investigation of its influence on the bubble absorption process. The results represented that the incorporation influence of the nanoferrofluid and the external magnetic field considerably improved ammonia-water bubble absorption.

On the other side, problems with acetone include high volatility, flammability, low density, low viscosity, difficulty in adding ionic components (insolubility), no concept of pH, difficulty manipulating the external continuous phase, and hardening of many plastics used in beakers and tubing. But a sufficient dispersivity of acetone or any medium has acetone as a component in it for many

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oxides, which has been documented in the literature [12]. Where the magnetite particles are scattered in acetone and detached by filtrate easily [13]. In Ajib and Karno's study [14], the dissolubility experiments of acetone/$ZnBr_2$ working solution have shown that with a concentration of 30% of $ZnBr_2$ almost all of the acetone has been vaporized at experimental conditions 50ºC and a 750 mbar. Other samples have been investigated with different experiment conditions, it had satisfying results too. Therefore, this working solution is attractive for being a cooling mixture fluid in ARSs driven by different low temperatures sources.

The main objective of this study is to perform the visual inspection of nanoparticle stability and thermophysical analysis for the mixture of working solutions of (Fe_3O_4–acetone/$ZnBr_2$). Then examining its efficiency characteristics as a nanoferrofluid for use in ARS. In literature, only one experimental investigation has been performed to determine the thermophysical properties of the suggested acetone/$ZnBr_2$ solution, [14]. And just one more researcher studied the thermophysical properties of the suggested working solutions as nanofluids by using ZnO and graphene in two separate investigations as suspension nanoparticles, [15-16]. At the same time, no previous study has tested the nanoferrofluid based acetone/$ZnBr_2$ as it is performed in this work. So this work can be considered an investigation of a new nanoferrofluid and a continuation of the previous works.

Thermophysical analyses (density, specific heat capacity, dynamic viscosity, and thermal conductivity) were achieved to give a base for finding the optimal mixture of working solution of (Fe_3O_4–acetone/$ZnBr_2$). Also, it was examined its efficiency characteristics as a nanoferrofluid as a candidate binary solution for use in ARS driven by different low temperatures sources. It shows also an investigation of (Fe_3O_4-acetone/$ZnBr_2$) as a nanoferrofluid including the preparation, stability, structure, and properties. The thermophysical properties are tested at a liquid saturated case at the atmospheric pressure or vapor pressure. The uncertainties for the experiments were relying on the tested properties and the used devices.

2. Materials and Methods

2.1 Preparation

The elaboration of nanofluid needs some distinctive ways for less agglomeration and sedimentation problems. Basically, nanofluid is created by dispersing metal, metal oxides, non-metals in base fluids such as water and brine. Water and brine-based nanofluids elaboration have been described in the literature such as (Saidur et al. [17]; Kumar et al. [18]; Lin et al. [19]; Nourafkana et al. [8]). Fundamentally, the preparation of nanofluid is summarized in two processes: a single-step process and a two-step process. These can be performed by utilizing the chemical method or mechanical one. In the single-step, process nanoparticles are synthesized and dispersed in the liquid simultaneously. In the two-step, process nanoparticles are synthesized separately then dispersed in a base fluid and maintaining its stability. The one-step process is better in terms of nanoparticle agglomeration. But for the two-step process is easier for obtaining, especially when the researchers who work on this project have no enough experience in the chemistry field.

The nanoferrofluid (Fe_3O_4-acetone/$ZnBr_2$) is prepared with different fraction mass concentrations (40 wt.% - 65 wt.% of $ZnBr_2$, 0.05 wt.% - 0.2 wt.% of Fe_3O_4) using the two-step process. The experiments are designed to see how effects of various concentrations of the nanoferrofluid on the thermophysical properties. Then obtaining the optimal concentration of the nanoferrofluid components will be candidates for use in ARS depending on the best physical properties. The most effective approach for enhancing nanoparticle suspension is prepared as follows; the nanoferrofluid samples are left in an ultrasonic device (bath type, 40 kHz, 160 W, model HY- 6 Lt D) produced by HI TEKNOLOJI LTD for 2 hours [20].

During preparing the based-fluid working solution heat is emitted from the mixture because of the dissolve reaction, which may reach 65°C depending on added $ZnBr_2$ concentration to the acetone. So the mixture is cooled down in certain time periods during mixing because the heating of metal particles would lead to the evaporation of refrigerant in the mixture [15]. In order to understand the dispersion characteristics of the Fe_3O_4 nanoparticles in the solutions for the experiments, a Scanning Electron Microscope (SEM) analysis of the nanoferrofluid samples is carried out. The tested nanoferrofluid samples with different concentrations after the sonication process in a visible way using sedimentation by gravitation over time were also compared with a non-sonicated sample. Then testing the density, heat capacity, thermal conductivity, and viscosity of different concentrations of base fluid and nanoferrofluid.

Sources of the materials: (Acetone, $ZnBr_2$, pure nanoparticles Fe_3O_4 99% (50-100 nm) particle size (SEM), 97% trace metals basis) were ordered from RASAL KIMYA Company Ltd.

2.2. Stability

1) Visual Inspection of Acetone/$ZnBr_2$ Base Fluid

Mohammed et al. [15] had found that the heat or light (or both of them) has an effect on changing the color of acetone/$ZnBr_2$ and becomes brown over time. However, in the current study, it is found that even in a dark medium (dimming by using aluminum paper) with in-room temperature (25 °C), the color of the solution changes to brown over time as is shown in Figure 1. This is due to the degradation of $ZnBr_2$ forming bromine. Where it was noticed that the samples that have higher concentrations (more than 50% of $ZnBr_2$) turned brown obviously.

2) Dispersion Characteristics of Fe_3O_4 Nanoparticles in The Base Fluid

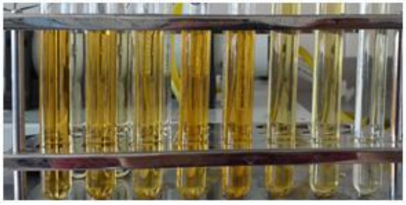
In order to overcome the long-range magnetic forces between the nanoparticles in the base fluid, a sonication process was performed experimentally on the nanoferrofluid (acetone – $ZnBr_2$/ Fe_3O_4, 50% $ZnBr_2$ of base fluid) samples with Fe_3O_4 nanoparticle concentrations of 0.05, 0.1, 0.15, and 0.2 wt.%. The nanoferrofluid samples are left in an ultrasonic device (bath type, 40 kHz, 160 W, model HY- 6 Lt D) produced by HI TEKNOLOJI LTD for 2 hours [20]. After several pre-experiments performed on different samples that had different concentrations of nanoparticles in the used ultrasonic device with various sonication process periods, it can be inferred the sonication process time on the nanoferrofluid has no significant influence on the suspension stability.

3) Visual Inspection of Stability

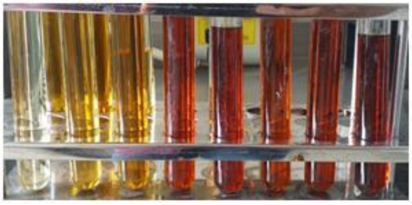
Figure 2 illustrates nanoferrofluid suspensions with several concentrations of Fe_3O_4. Tube number 5 was prepared without sonication and with 0.1 wt.% of Fe_3O_4

concentration. It can be seen from Figure 2a that the sonication process has an obvious influence on the samples comparing with tube 5 (non-sonicated sample). Sonicated samples have less agglomeration and the upper section of tube 5 is, somewhat, more net than the same section of others over time. On the other hand, from Figures 2b, 2c, and 2d, it can be realized that although there is an increasing trend to produce bigger agglomerations in samples that have a greater concentration of nanoparticles, there is no obvious difference in sedimentation in each solution under natural gravity. These results elucidate that the nanoferrofluid has good dispersion and weak stability characteristics. That pushes us to use an additional method by applying a magnetic vortex to get good stability when this nanoferrofluid is applied in ARS.

(After mixing directly)



(After 4 days)



(After 16 days)

Figure 1. Color changes of acetone/$ZnBr_2$ solution samples with several concentrations by the time.

4) Examination of The Chemical Surfactant Factors Effects

Three chemical surfactant factors (poly vinyl pyrrolidone (PVP), sodium dodecyl sulfate (SDS), and polyvinyl alcohol (PVA)) were examined in a try to enhance the suspension nanoparticles stability. The sample used in this test was a solution with a 0.1 wt.% concentration of nanoparticles that were prepared with the same by adding 1 wt.% of the three surfactants separately. Figure 3 shows that these types of surfactants have no useful effect on the dispersion of the nanoparticles over time. Where directly after the sonication process particles start agglomerating. Also, the surfactants cannot dissolve and start to collect on the top of the solution. Over time these effects become clearer.

5) Scanning Electron Microscope (SEM) Analysis

The surface morphology and particle sizes of the samples were carried out using a high-resolution SEM (JEOL-JSM-7001F) type which exists in Black Sea Advanced Technology Research and Application Center Laboratories (KITAM) operating at 15 kV. Figure 4 shows SEM images of Fe_3O_4 nanoparticles of 0.1 wt.% mass concentration nanoferrofluid. The images display no clear agglomeration, which shows the Fe_3O_4 nanoparticles have almost a typical and random distribution situation in the liquid. This distribution occurred after sonication because of just the nanoparticles' thermal motion.

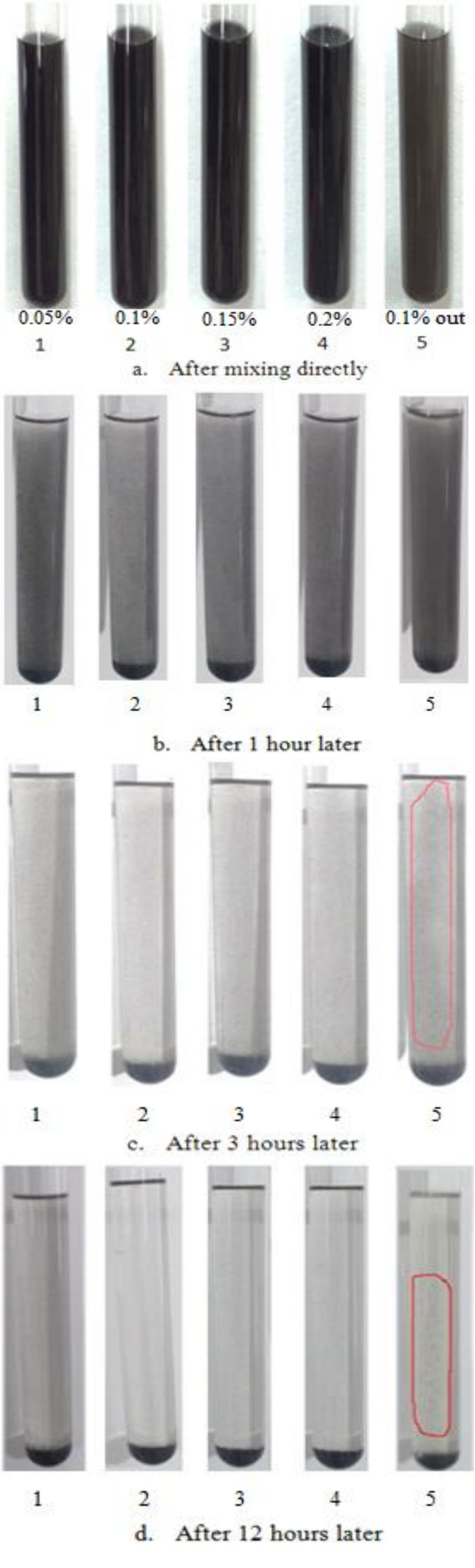
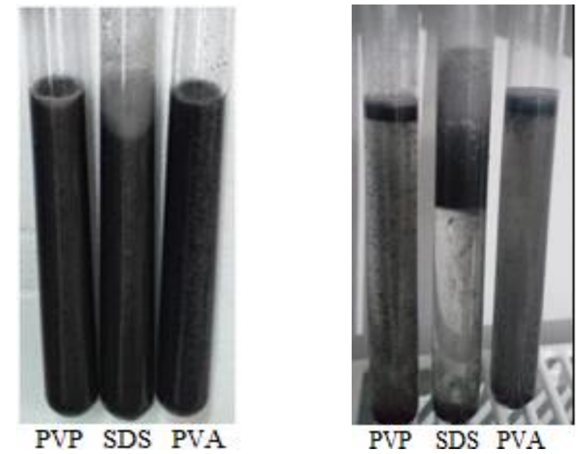
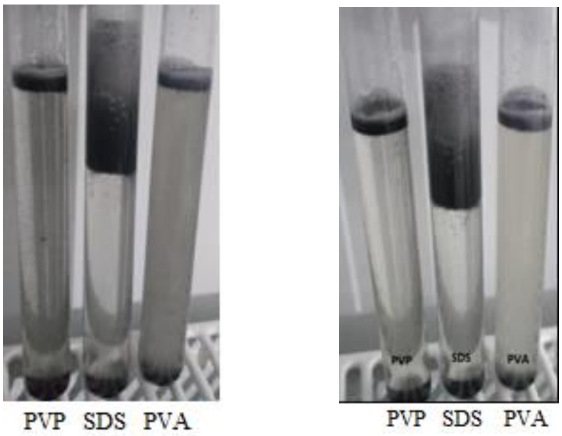


Figure 2. Sedimentation and agglomeration of varies concentrations of Fe_3O_4 nanoparticles in acetone/$ZnBr_2$ base fluid. 1–0.05 wt.%; 2–0.1 wt.%; 3–0.15 wt.%; 4–0.2 wt.% (with 2 h sonication). 5–0,1 wt.% (without sonication)



(Directly after sonication) (1 hours later)



(3 hours later) (12 hours later)

Figure 3. Effects of using different surfactants on the stability of nanoferrofluid (Fe_3O_4-acetone/$ZnBr_2$) over time.

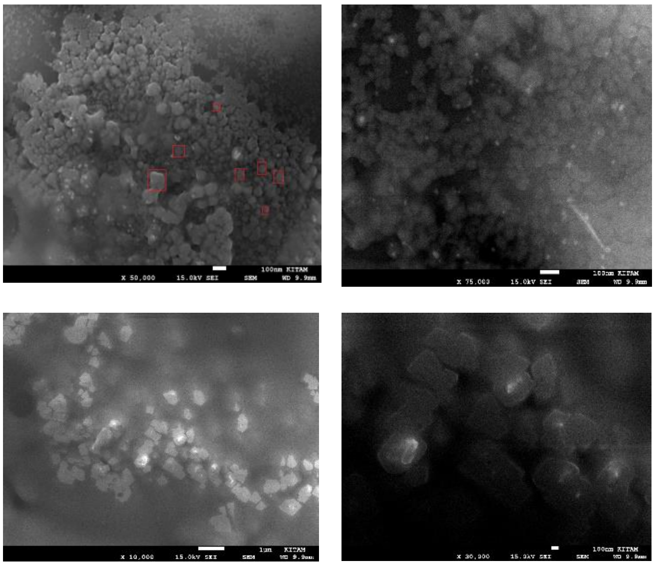


Figure 4. Various SEM images of Fe_3O_4 nanoparticles suspension in the acetone/$ZnBr_2$ base fluid.

3. Results and Discussion

3.1 Thermophysical Properties

1) Density

The measured density of several acetone/$ZnBr_2$ concentrations at 25°C in this study was significantly similar to those investigated by Mohammed et al. [15] and dissimilar to which was investigated by Ajib and Karno [14]. Because of the increase of the solution volume when the salt ($ZnBr_2$) is added and dissolved in the solvent (acetone) which was not taken into account in Ajib and Karno study [14]. During the experiment by taking the volumetric flask error (0.5 to 1%) into consideration and depending on the volumes used (10 ml sample), Table 1 represents the variation among the measured densities of several acetone/$ZnBr_2$ concentrations as found experimentally in this study and the literature. Figure 5 shows an increase in the density of the nanoferrofluid from 1.366 to 1.606 g cm^{-3} with an increase in concentration of the nanoparticle from 0.0 to 0.2 wt.%. And also, almost identical values of the theoretical density (which are set by Equation 1) with experimental ones [15].

$$\rho_{tot} = \frac{1}{\frac{X_s}{\rho_s} + \frac{X_p}{\rho_p}} \tag{1}$$

Where the subscripts *tot*, *s*, and *p* indicate nanoferrofluid, base-solution and nanoparticles respectively. ρ and X indicate the density and mass fraction respectively. ρ_p =5 g cm^{-3} [21].

Table 1. The density of acetone/$ZnBr_2$ with various concentrations of acetone as measured experimentally in this study at 25 °C and in similar studies.

$ZnBr_2$ Concentration	Reported density in [14] (g cm^{-3})	Reported density in [15] (g cm^{-3})	Measured density of this study (g cm^{-3})
40 %	1.291	1.164	1.161
45 %	1.420	1.228	1.230
50 %	1.581	1.360	1.366
55 %	1.778	1.426	1.431
60 %	2.013	1.550	1.553
65 %	2.292	1.662	1.663

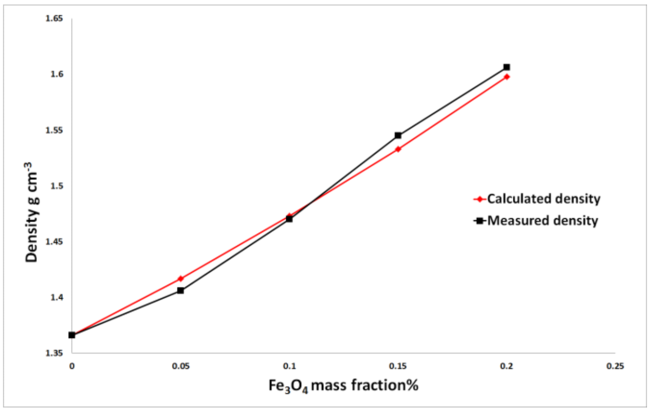


Figure 5. Changes of the nanoferrofluid density with different Fe_3O_4 nanoparticles mass fraction, experimentally and theoretically

2) Viscosity

The viscosity of several concentrations of acetone/$ZnBr_2$ was measured at 25°C, using Thermo Scientific™ HAAKE™ MARS™ 40 Rheometer, [22]. The results of this test have no substantial difference from those measured in literature [14] and [15]. On the other hand, it can be realized from Figure 6 that a major increase in viscosity happened from 3.384 to 9.206 mPa s with increasing concentration of Fe_3O_4 from 0 to 0.2 wt.%. Hence, the nanoferrofluid has a high viscosity comparing with its base fluid. And also has a high viscosity comparing with another kind of nanofluid with the same base fluid [15]. The viscosity measurements were repeated at least 3 times. The measurement error at the given temperature was less than 1%. It is assumed that nanoferrofluid has a single-phase liquid.

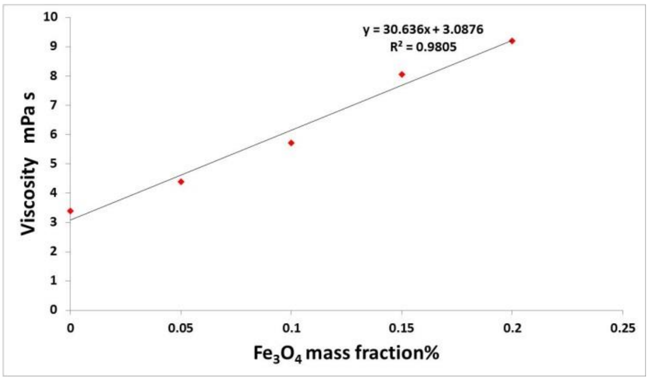


Figure 6. Changes of the nanoferrofluid density with different Fe_3O_4 nanoparticles mass fraction, experimentally and theoretically.

3) Specific Heat Capacity at Constant Pressure

The specific heat capacity of several concentrations of acetone/$ZnBr_2$ base fluid was measured using KD2 Pro Thermal Properties Analyzer, Decagon Devices, Inc. [23]. Figure 7 illustrates the specific heat capacity of the Fe_3O_4-acetone/$ZnBr_2$ (50 wt.% $ZnBr_2$) with various concentrations of Fe_3O_4 indicated experimentally and theoretically at 25 °C. While there are several methods for assessing the heat capacities of pure liquids, very few set correlations have been proposed for mixtures. Furthermore, since there is no model obtainable to calculate the specific heat capacity of nanofluids, a simple method (which is set by Equation 2) dependent on the corresponding state principle is proposed for the calculation of the heat capacities of liquid mixtures [24-25].

$$c_{p_{tot}} = X_s c_{p_s} + X_p c_{p_p} \tag{2}$$

Where c_p is the specific heat capacity.

It can be realized from Figure 7 that the specific heat capacity decrease with increasing the particles (Fe_3O_4) concentration. It is a logical consequence because of the low heat capacity value of the nanoparticles (0.653 J g^{-1} K^{-1})compared with the specific heat capacity of acetone/$ZnBr_2$ (1.140 J g^{-1} K^{-1}) and the higher concentration, the higher agglomerations. More agglomeration causes bigger particles in the nanoferrofluid which drops the total heat capacity and that is also what was mentioned in the literature with other kinds of nanofluids [26-27]. As a result of the analysis, there was an excellent suspension of nanoparticles in this work. Also, there is no major decline in tested values to the computational values of the heat capacities comparing with other kinds of nanoparticles that have been studied before in the literature.

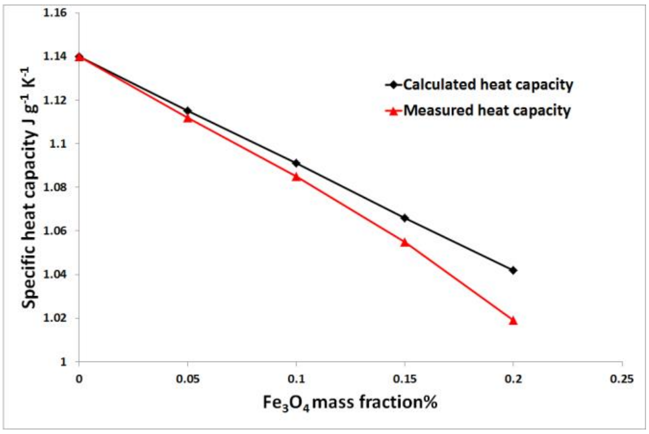


Figure 7. Changes of the nanoferrofluid specific heat capacity with different Fe_3O_4 mass fractions, experimentally and theoretically.

3.2 Thermal Conductivity

Thermal conductivity is a very important property in our study. Because it can be used to illustrate the enhancement of heat transfer in the generator of ARS by using the nanoferrofluid instead of the conventional working solution, which is the main aim of this work. Ajib and Karno [14] have not studied this property in their research. Thermal conductivity of acetone/$ZnBr_2$ with several concentrations of $ZnBr_2$ in the solution (40–65 *wt.*%) was tested using KD2 Pro Thermal Properties Analyzer, Decagon Devices, Inc. [23]. The results of the thermal conductivity measurements of acetone/$ZnBr_2$ which were obtained in this study are significantly similar to those were by Mohammed et al. [15]. After performing 2 hours sonication process, the nanoferrofluid samples (The base fluid is acetone/$ZnBr_2$ with 50 *wt.*% of salt) with different concentrations of nanoparticles (0.05, 0.1, 0,15, and 0.2 *wt.*% of Fe_3O_4) experimentally and theoretically were studied.

The effective thermal conductivity can be calculated theoretically by

$$k_{\mathrm{Maxwell}} = \left[\frac{2k_s + k_p + 2\varphi(k_p - k_s)}{2k_s + k_p - \varphi(k_p - k_s)}\right] k_s \tag{3}$$

Where φ refer to the volume fraction. $k_p=0.61\ W\ m^{-1}\ K^{-1}$ at $25°C$ [28].

It is known as the classical Maxwell's equation [29]. Later, a lot of researchers tried to improve this model by correlating it to their experimental studies. Thus, they proposed predictive models for thermal conductivity [30-31]. However, their model is complex and there are different experimental parameters, including the fractal dimension and the ratio of the minimum to a maximum diameter of nanoparticles. Also, they do not have satisfying results in such the current study. As is shown in Table 2, he thermal conductivity increases from 0.150 to 0.167 $W\ m^{-1}\ K^{-1}$, when the concentration of the Fe_3O_4 nanoparticles increases from 0 to 0.2 *wt.*%, also the enhancement of thermal conductivity reaches 10.179 % at 0.2 *wt.*%.

Table 2. The thermal conductivity of the Fe_3O_4-acetone/$ZnBr_2$ (50 wt.% $ZnBr_2$) with various concentrations of Fe_3O_4 indicated experimentally and theoretically in this study at 25 °C.

Fe_3O_4 nanoparticles Concentration (*wt*.%)	Calculated thermal conductivity $k_{Maxwell}$ ($W\ m^{-1}\ K^{-1}$)	Measured thermal conductivity ($W\ m^{-1}\ K^{-1}$)	Enhancement %
0.00	0.15000	0.150	-
0.05	0.15022	0.157	4.458
0.1	0.15045	0.161	6.832
0.15	0.15068	0.164	8.536
0.2	0.15091	0.167	10.179

The maximum values of the uncertainty had a considerable amount during the preparation experiments (due to the used analyzer). Later, we overcame this by performing the measurements for 3 same samples from every studied concentration and more than 15 times for every sample of them at the same ambient conditions. Finally, the measurement error at a given temperature was less than 1.5 %. Also, It can be realized from Table 2 the difference in thermal conductivity between the theoretical (depending on the Maxwell model) and experimental values which are related to the concentration of Fe_3O_4 nanoparticles. Where although the high viscosity value comparing with base fluid the Fe_3O_4-acetone/$ZnBr_2$ which weakens the Brownian motion of suspended nanoparticles, the Maxwell equation does not give a significant prediction on the thermal conductivity of nanoferrofluid without Brownian motion of suspended nanoparticles. Moreover, the conduction part of the prediction model cannot be obtained from Maxwell prediction [31].

4. Conclusion

This study presented an investigation of Fe_3O_4-acetone/$ZnBr_2$ as a nanoferrofluid including the preparation, stability, structure, and properties. The reason behind choosing Fe_3O_4 nanoparticles is the ability of nanoferrofluid to be used under influence of an external magnetic field which causes a macro-magnetic force and a magnetic–thermal effect in the nanoparticles, which is not available with conventional nanofluid. The results are represented as follows:

1. It was found that even in a dark medium with in-room heat (25 °C), the color of the base fluid changes over time because of the degradation of $ZnBr_2$ forming bromine.
2. The visual inspection analysis elucidates that the Fe_3O_4 nanoferrofluid of the base fluid has good dispersion and weak stability characteristics.
3. Using different surfactants had a bad effect on suspension nanoparticles' stability.
4. SEM analysis showed a typical and random distribution situation of the nanoparticles in the base fluid.
5. By increasing nanoparticle concentration, the density increase, heat capacity decrease, as expected, and viscosity had a significant increase.
6. The results of the thermal conductivity measurements of the base fluid which were obtained in this study are significantly similar to those were in the literature.
7. Maxwell's model (as a theoretical method to find the thermal conductivity) did not give a significant prediction on the thermal conductivity of nanoferrofluid without the Brownian motion of suspended nanoparticles comparing with the experimental results.
8. The enhancement of thermal conductivity reaches 10.179 % at 0.2 *wt*.% nanoparticle concentration.

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