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Effects of Surfactants on the Stability of Nickel Ferrite/Water Nanofluid

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ABSTRACT

Nanofluid stability is a critical factor for the effective application of nanofluids in various fields. One simple and effective method to enhance nanofluid stability is through the addition of surfactants. This study examines the effect of different surfactants on the stability of nickel ferrite ($NiFe_2O_4$)/water nanofluid. The nanofluids were synthesis using the two-step method, and the surfactants investigated inculded oleic acid, polyethylene glycol 400, tetrabutylammonium bromide, gum arabic, and citric acid. Different concentrations for each surfactant were tested by adjusting the nanoparticles-to-surfactant ratio. The suspension stability was evaluated through visual observation, Zeta potential measurements, and thermal conductivity analysis. The most stable $NiFe_2O_4$/water nanofluid was achieved using citric acid surfactant, with a nanoparticles-to-surfactant volume ratio of 1:0.25, a Zeta potential value of -35.0 mV and an average thermal conductivity of 0.585 ± 0.007 W/m·K. The results of this study are important for developing nanofluid and magnetic nanofluid systems with optimum conductive heat transfer performance.

1. INTRODUCTION

Modern human life relies heavily on machines designed to improve comfort and efficiency. However, during operation, these machines generate significant amount of heat that must be dissipated to prevent increased system pressure due to inadequate cooling. Alternatively, the excess heat produced during system operation can be stored using a thermal energy storage (TES) system for future use. This can be achieved through either sensible heat storage or latent heat storage (Alva et al., 2018). Heat transfer fluids (HTFs) are commonly employed in heat transfer devices, primarily to dissipate excess heat or to transfer it to TES system for specific applications (Okonkwo et al., 2021). The heat transfer efficiency of HTFs primarily depends on their thermal conductivity. Enhancing the thermal conductivity of HTFs is therefore essential for improving the overall performance of heat transfer systems (Guo, 2020).

For several decades, conventional fluids such as water, ethylene glycol, and oil have been used as HTFs (Huminic & Huminic, 2016). The performance of these conventional fluids can be significantly enhanced by incorporating materials with high thermal conductivity, particularly in nanometer range (1 to 100 nm), commonly referred to as nanofluid (Younes et al., 2022). Nanofluids are suspensions or colloidal systems consisting of conventional fluids containing nanoparticles of metals or metal oxides, such as Ag, Cu, Zn, CuO, ZnO, MgO, Al_2O_3, SiO_2, TiO_2, etc., or carbon-based materials such as graphite, graphene, or carbon nanotubes (Ahmadi et al., 2018; Ataei et al., 2020; Yadav et al., 2022; Jung & Park, 2021). The dispersion of a small quantity of these nanoparticles can significantly improve the thermal properties of conventional fluids.

An advanced development in nanofluids is the introduction of ferrofluids, which are suspensions of HTFs containing iron-based nanoparticles. Further advancements include magnetic nanofluids, which incorporate other types of magnetic nanoparticles (Kumar & Subudhi, 2018), as the thermal conductivity of these systems can be further enhanced by applying an external magnetic field (Selim et al., 2023; Doganay et al., 2019). Among the common magnetic nanoparticles used in these systems include Fe_2O_3, Fe_3O_4, Co_3O_4, or spinel ferrites, with the general formula MFe_2O_4, where M represents divalent transition metal ions, such as Ni, Co, Zn, Cu, or Mn (Amiri et al., 2019). However, one important aspect for the application of nanofluids, ferrofluids, or magnetic nanofluid systems is their stability, as the instability may lead to sedimentation or agglomeration, both of which reduce the effectiveness of the system. A simple method to improve the stability of nanofluids is the addition of surfactants. Surfactants prevent agglomeration by coating the nanoparticles, thereby reducing sedimentation (Wang et al, 2023).

The stabilization methods for nanofluids using surfactants are typically categorized into three main types: electrostatic, steric, and electrosteric stabilization (Chakraborty & Panigrahi, 2020). Electrostatic stabilization mostly uses ionic compounds, and in this case the nanoparticles acquire surface charges either through ionization or by adsorbing ions from the dispersing medium. Nanofluid stability is achieved when the electrostatic repulsion between similarly charged particles prevents agglomeration caused by van der Waals attraction forces (Wang et al., 2023; Mehta et al., 2022). Steric stabilization of nanofluids involves the addition of non-ionic or polymer molecules. These polymer molecules form a steric hindrance, preventing nanoparticles from sticking together. When using polymers as surfactants, the polymer chains adsorb onto the nanoparticle surfaces, restricting the free movement of the base fluid (Chakraborty & Panigrahi, 2020). Electrosteric stabilization is a combination of both electrostatic and steric stabilization, utilizing both ionic and non-ionic surfactants.

Various techniques can be employed to evaluate nanofluid stability, including visual inspection, centrifugation, Zeta potential measurement, Uv-vis spectral analysis, transmission electron microscopy (TEM), and dynamic light scattering (DLS). Additionally, changes in physical properties, such as thermal conductivity and viscosity, can also be used to assess the stability of nanofluids (Chakraborty & Panigrahi, 2020). In particular, nanofluids with a Zeta potential (ζ) higher than +60 mV or less than -60 mV exhibit excellent stability, nanofluids with ζ values between ±30 mV and ±60 mV demonstrates good stability, whereas ζ values between +30 mV and -30 mV indicate unstable nanofluid (Chakraborty & Panigrahi, 2020).

An experimental study by Karimi et al. in 2015 revealed that the thermal conductivity of $NiFe_2O_4$/water nanofluids was lower than of Fe_3O_4/water nanofluids at relatively low nanoparticle concentrations. However, this trend reversed at higher concentrations, specifically above 1% vol (Karimi et al., 2015). These results raise questions about the role of the nanoparticle in the NFs, since the viscosity of nanofluids also increases with increasing particle concentration in addition to enhancing thermal conductivity, and the viscosity of NFs ultimately affects their thermal conductivity (Younes et al., 2022).

Furthermore, in an external magnetic field, the enhancement of thermal conductivity of the MNF system is determined by the saturation magnetization of the nanoparticles (Katiyar et al., 2016). We note that the surfactant used to stabilize the suspension significantly influences the thermal conductivity value of the nanofluid. While many types of surfactants have been studied for Fe_3O_4-based nanofluids, there is little data for spinel ferrites, particularly nickel ferrite ($NiFe_2O_4$) (Wang et al., 2021; Selim et al., 2023; Doganay et al., 2019). Thus far, surfactants used for $NiFe_2O_4$/water nanofluids have been primarily restricted to tetramethylammonium hydroxide (TMAH) and sodium dodecylbenzene sulfonate (SDBS) (Karimi et al., 2015; Yılmaz et al., 2023).

This paper aims to investigate the effects of various surfactants on the stability of $NiFe_2O_4$/water nanofluids. Compared to other types of HTFs, water exhibits excellent thermal properties, making it a common choice for conventional HTFs (Mehta et al., 2022). In this research, we utilized five surfactants: oleic acid (OA), polyethylene glycol 400 (PEG 400), tetrabutylammonium bromide (TBAB), gum arabic (GA), and citric acid (CA). For each surfactant, we applied varying amounts by adjusting the volume ratio between the nanoparticles and the surfactant. The stability of the nanofluid suspensions was assessed qualitatively by visual observation and quantitatively by measuring the Zeta potential and thermal conductivity for the most stable nanofluid.

2. METHOD

2.1 Materials

Nickel ferrite ($NiFe_2O_4$) nanoparticles were obtained from MTI Corporation with a purity of 99.5% (MTI Corporation, 2023). Tetrabutylammonium bromide was sourced from Wuhu Nuowei Chemistry with a purity of 98% (Wuhu Nuowei Chemistry, 2023). Technical oleic acid, polyethylene glycol 400, gum arabic, citric acid, and distilled water were procured from Subur Kimia Jaya. Table 1 summarizes the solubility of each surfactant in water and their classifications.

Table 1 Solubility of various surfactants in water and its classification.

Surfactants	Solubility in water	Classification	Reff
Oleic acid ($C_{18}H_{34}O_2$)	-	Non-ionic	(PubChem, 2024)
Citric acid ($C_6H_8O_7$)	1174g/L (10°C), 1809g/L (30°C), 3825g/L (80°C).	Ionic	(Pubchem , 2024)
Polyethylene glycol 400 ($C_{2n}H_{4n+2}O_{n+1}$)	670 mg/mL (20 °C)	Non-ionic	(PubChem, 2024)
Tetrabutylammonium bromide ($C_{16}H_{36}NBr$)	600 g/L (20 ºC)	Ionic	(PubChem, 2024)
Gum arabic	Solute (there is no quantitative data yet)	Non-ionic	(PubChem, 2024)

2.2 Nanofluid Preparation

The synthesis of nanofluid applied the two-step method (Younes et al., 2022), with the nanofluid volume fraction (ϕ) calculated using Equation (1):

$$\phi(\text{vol.}\%) = \frac{[m_{np}/\rho_{np}]}{[m_{np}/\rho_{np}] + [m_{bf}/\rho_{bf}]} \times 100 \quad (1)$$

where m_{np} and ρ_{np} are the mass and density of the nanoparticles, respectively, while m_{bf} and ρ_{bf} are the mass and density of base fluid. For calculation we used ρ_{np}= 5.368 g/cm³ for the density of $NiFe_2O_4$ nanoparticle (MTI corporation) and ρ_{bf} = 0.997 g/cm³ for the density of water. In this study we use ϕ = 0.5 vol.%.

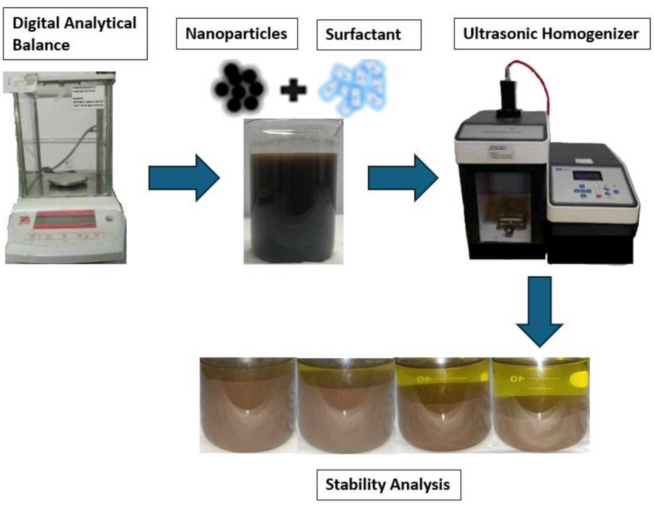


Figure 1. Nanofluid synthesis with the two-step method and visual observation to specify the nanofluid stability.

We prepared different nanoparticles to surfactant ratios for each surfactant, namely 1:1 and 1:2 for oleic acid; 1:0.125 and 1:0.25 for PEG 400; 1:0.25, 1:0.5, 1:0.75 and 1:1 for tetrabutylammonium bromide; 1:0.25, 1:0.5, 1:0.75, 1:1 and 1:2 for gum arabic; and 1:0.25, 1:0.375, 1:1, 1:2 and 1:4 for citric acid. The nanoparticles and surfactants were added to distilled water and stirred until a homogeneous mixture was obtained. The suspension was then ultrasonicated for a total of 3 hours at 40% power. The obtained suspension was then visually observed to evaluate its stability, as illustrated in Figure 1.

2.3 Nanofluid Stability

The stability of the nanofluids was observed at 10-minute intervals over a maximum duration of 1 hour. The most stable nanofluid was subsequently evaluated through Zeta potential measurements and time-dependent thermal conductivity analysis (Ibna & Bodius, 2020). Zeta potential measurement were conducted using a Horiba SZ-100 Nano Particle Analyzer at the Nanosains and Nanotechnology Research Center (PPNN), ITB. The thermal conductivity was measured using a KD2 Pro Thermal Properties Analyzer, which operates based on the transient hot wire method (Healy et al., 1976).

3. RESULTS AND DISCUSSION

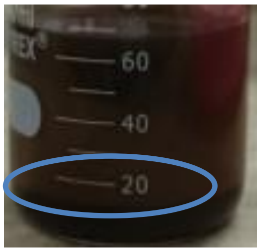
3.1 Qualitative Observation for Nanofluid Stability

Table 2 presents the stability test results for $NiFe_2O_4$/water nanofluids stabilized with oleic acid (OA) surfactant at $NiFe_2O_4$-to-OA volume ratios of 1:1 and 1:2. The data indicate that the $NiFe_2O_4$/water/OA nanofluid did not achieve homogeneous dispersion, as evidenced by the sedimentation and agglomeration of the $NiFe_2O_4$ nanoparticles. The addition of oleic acid to the $NiFe_2O_4$/water nanofluid failed to enhance its stability. This instability may be attributed to the incomplete dispersion of oleic acid in water, which likely resulted in an ineffective coating of the nanoparticles. Consequently, the nanoparticles were prone to agglomeration and sedimentation. Figure 2 illustrates the agglomeration of the nanoparticles at the bottom of the fluids.

Table 2. Stability observation o $NiFe_2O_4$/water nanofluids tabilized with oleic acid (OA) surfactant

$NiFe_2O_4$: OA ratio	Observation time (min.)				
	1	5	10	15	30
1:1					
1:2					

a)



b)

Figure 2. Agglomeration of $NiFe_2O_4$/water nanofluid using OA surfactant after 1 hour of observation for various $NiFe_2O_4$: OA ratio of (a) 1:1 and (b) 1:2.

Table 3 and Table 4 present the stability observations results for $NiFe_2O_4$/water nanofluids using PEG 400 and TBAB surfactants. The images reveal that the nanofluids failed to achieve homogeneous dispersion with either surfactant. Separation was observed within 5 minutes and became more pronounced over time.

Table 3. Stability observation of $NiFe_2O_4$/water nanofluids stabilized with PEG 400 surfactant

$NiFe_2O_4$: PEG 400 ratio	Observation time (min.)				
	1	5	10	15	30
1:0.125					
1:0.25					

Table 4. Stability observation for $NiFe_2O_4$/water nanofluids using TBAB surfactant

$NiFe_2O_4$: TBAB ratio	Observation time (min.)				
	1	5	10	15	30
1:0.25					
1:0.5					
1:0.75					
1:1					

Table 5 indicates the stability observation for $NiFe_2O_4$/water nanofluids using gum arabic (GA) surfactant. The images indicate that a relatively small amount of gum arabic could promote the homogeneous $NiFe_2O_4$/water nanofluid with significantly less sedimentation after a few minutes. Additional GA concentration reduced the sedimentation, and the sedimentation disappeared for a $NiFe_2O_4$-to-GA volume ratio of 1:2.

Table 6 presents the stability observation of $NiFe_2O_4$/water nanofluids stabilized with citric acid (CA) surfactant. The images shows that the ratio $NiFe_2O_4$:CA = 1:0.25 produced nanofluids with good stability, maintaining suspension even after several days.

Figure 3 provides a quantitative analysis separations for observations for the stability of the nanofluids. $NiFe_2O_4$/water nanofluids stabilized with PEG 400 surfactant exhibited significant separation (see Figure 3(a)). The addition of PEG 400 did not enhance the stability of the nanofluid, likely due to the inability of the nanoparticles to adsorb the polymer chains of PEG 400, thereby failing to achieve effective steric stabilization. Similarly, $NiFe_2O_4$/water nanofluids stabilized with TBAB surfactant also showed significant separation, as depicted in Figure3(b). However, increasing the concentration of TBAB reducedsuspension separation. As an ionic surfactant, TBAB stabilizes the nanofluid via electrostatic interactions, where nanoparticles adsorb surfactant ions to counteract agglomeration, thereby minimizing separation and sedimentation. Despite this, TBAB remained relatively ineffective, as the nanofluid was unable to achieve long-term stabilization.

Table 5. Stability observation of $NiFe_2O_4$/water nanofluids stabilized with gum arabic (GA) surfactant

$NiFe_2O_4$: GA ratio	**Observation time (min.)**				
	1	**10**	**15**	**30**	**60**
1:0.25					
1:0.5					
1:0.75					
1:1					
1:2					

Table 6. Stability observation of $NiFe_2O_4$/water nanofluids stabilized with citric acid (CA) surfactant

NiFe2O4: CA ratio	**Observation time (min.)**				
	1	**10**	**15**	**30**	**60**
1:0.25					
1:0.375					
1:1					
1:2					
1:4					

In contrast, citric acid, a smaller ionic surfactant, demonstrated greater effectiveness in stabilizing $NiFe_2O_4$/water nanofluids (Figure 3(c)). However, increasing the citric acid concentration led to accelerated separation of the nanofluid. A nanoparticles-to-CA ratio of 1:0.25 yielded the best stability among all tested compositions, maintaining a well-dispersed suspension for an extended period.

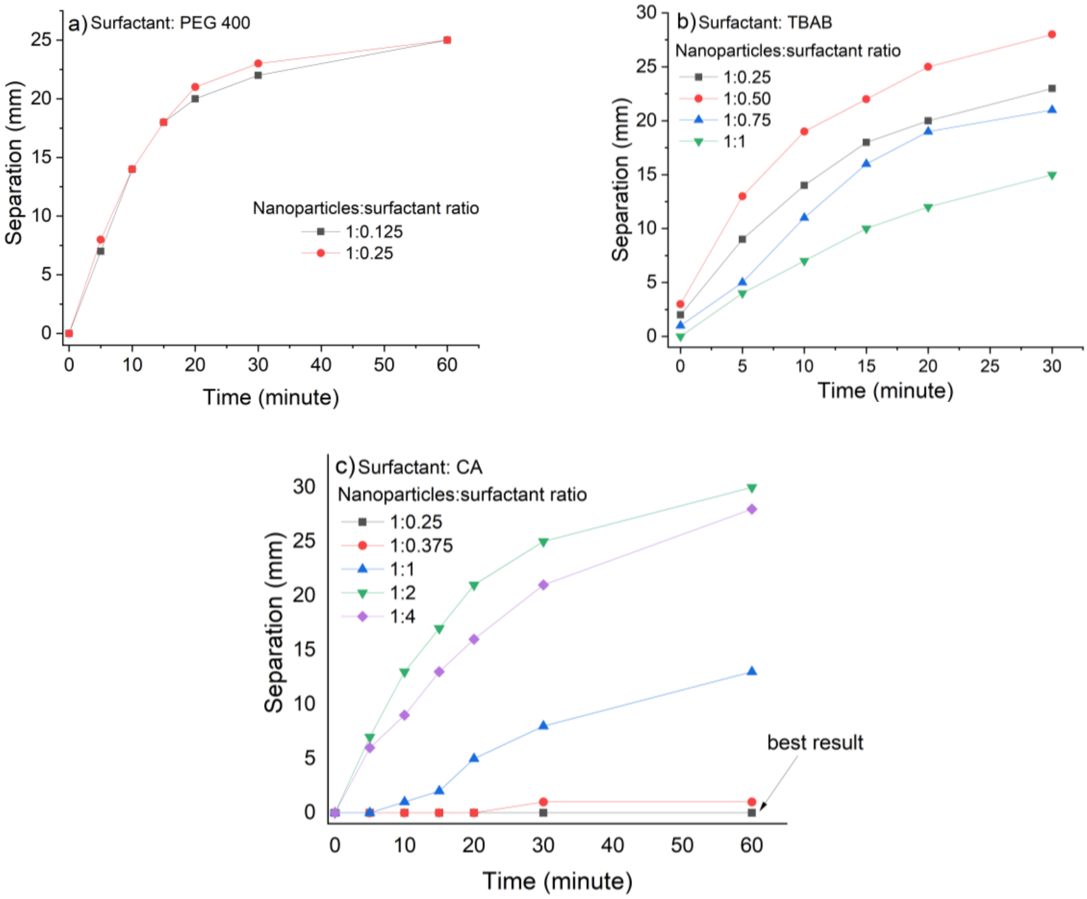


Figure 3. Separation observations of $NiFe_2O_4$/water nanofluid with different types of surfactants: a) PEG 400, b) TBAB, and c) CA.

Generally, the separation profiles shown in Figure 3 exhibit a similar trend across all types of surfactants, characterized by an initially high separation rate over a relatively short period, followed by a gradual decrease in the separation rate over time. In the aqueous base, homogeneously dispersed $NiFe_2O_4$ nanoparticles undergo collisions due to Brownian motion. According to the Derjaguin-Landau-Verwey-Overbeek (DLVO) theory, attractive forces between nanoparticles dominate over repulsive forces during collisions (Zheng et al., 2021). These attractive forces bring the nanoparticles closer together, resulting in agglomeration and sedimentation as the effective particle size increases. Following this rapid sedimentation phase, the rate of nanoparticle concentration reduction decreases,and the nanofluid shows improved stability (Zhang et al., 2020).

3.2 Zeta Potential and Thermal Conductivity Measurements

Figure 4 presents the Zeta potential measurements for $NiFe_2O_4$/water nanofluid stabilized with citric acid (CA) surfactant at nanoparticles-to-surfactant ratios of 1:0.25 and 1:0.375. The Zeta potential results indicate good stability, consistent with visual observations.

To further validate the stability of the $NiFe_2O_4$/water nanofluid with a nanoparticles-to-CA ratio of 1:0.25, thermal conductivity measurements were conducted every 15 minutes at room temperature (26.5°C), as shown in Figure 5(a). Over a 60-minute interval, the thermal conductivity decreases slightly from approximately 0.60 W/m·K to 0.58 W/m·K, and remained relatively constant there after for overtwo hours. The average thermal conductivity was measured as 0.585 ± 0.007 W/m·K, suggesting that the nanoparticle suspension was well-dispersed, and experience minimal aggregation.

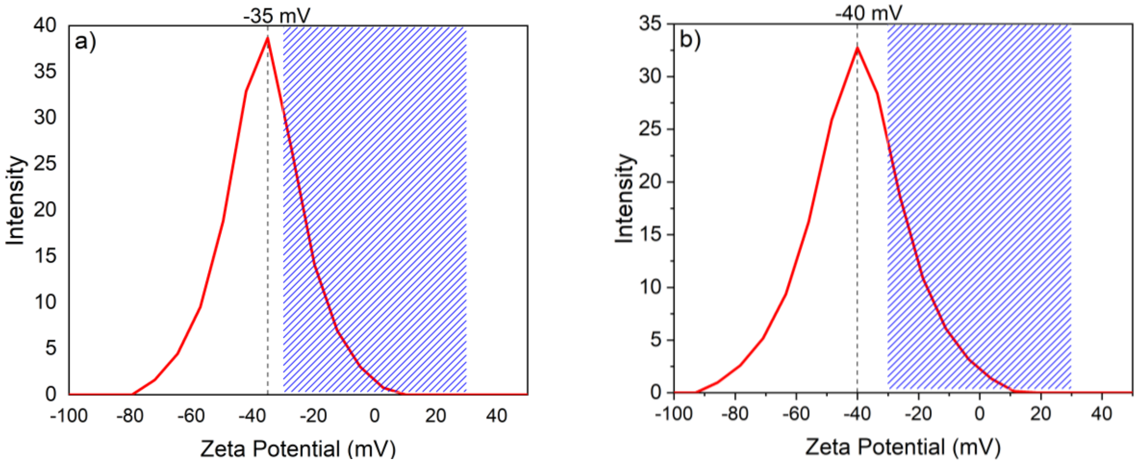


Figure 4. Zeta potential of $NiFe_2O_4$/water nanofluid using CA surfactant with nanoparticles-to-surfactant volume ratios of a) 1:0.25 and b) 1:0.375. The shaded area indicates the range of zeta potential values where the stability of the nanofluid is poor.

Figure 5(b) compares the thermal conductivity values of pure water, water with citric acid as the base fluid, and the most stable $NiFe_2O_4$/water nanofluid nanoparticles-to-CA ratio of 1:0.25). The results demonstrate a thermal conductivity enhancement of approximately 6% for the optimized nanofluid compared to the base fluids.

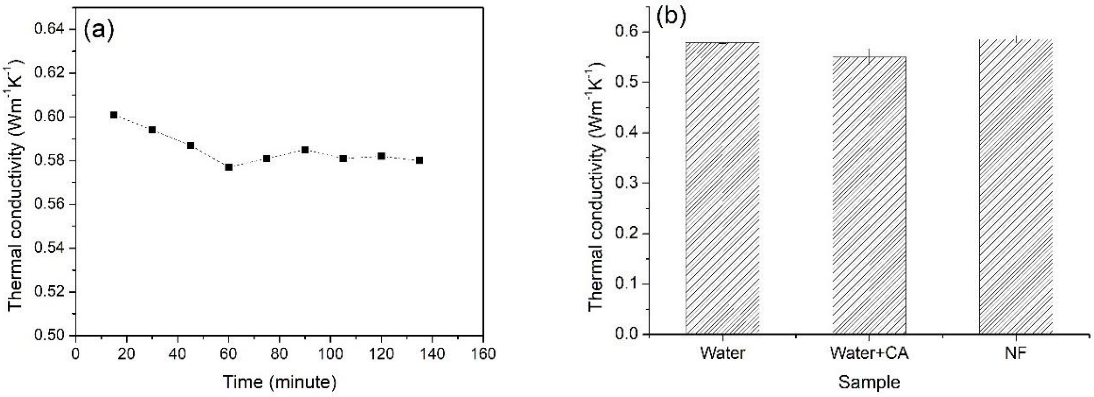


Figure 5. (a) Thermal conductivity of $NiFe_2O_4$/water nanofluid stabilized with citric acid (CA) surfactant at a nanoparticles-to-CA volume ratio of 1:0.25 over a period exceeding 2 hours. (b) Thermal conductivities of pure water, citric acid solution, and the best-obtained nanofluid.

We observe that the temperature of the nanofluid increases slightly during the ultrasonication process. This temperature rise likely creates favorable conditions for nanoparticle dispersion in the fluid by enhancing Brownian motion. In this context, the efficiency of nanoparticles collisions is directly related to temperature and inversely related to viscosity. Additionally, increasing the working temperature improves nanoparticle interaction energy (Wang et al., 2023).

Citric acid in water helps prevent particle agglomeration by generating free ions. These, nanoparticles adsorb the free ions, which improves electrostatic interactions between the particles and balancing dipolar and van der Waals attractions, ensuring that the particles remain dispersed in the fluid. Consequently, the NiFe2O4/water nanofluid, with relatively low concentrations of citric acid surfactant exhibits stability, preventing significant changes in thermal conductivity over time.

4. CONCLUSION

This paper presents an experimental study on the effect of surfactant type and concentration on the stability of (nickel ferrite) $NiFe_2O_4$/water nanofluid with a concentration of 0.5 vol.%. Various surfactants and nanoparticles-to-surfactant volume ratios were evaluated, including oleic acid (1:1 and 1:2), polyethylene glycol 400 (1:0.125 and 1:0.25), tetrabutylammonium bromide (1:0.25, 1:0.5, 1:0.75 and 1:1), gum arabic (1:0.25, 1:0.5, 1:0.75, 1:1 and 1:2), and citric acid (1:0.25, 1:0.375, 1:1, 1:2 and 1:4). Nanofluids stabilied with oleic acid, polyethylene glycol 400, and tetrabutylammonium bromide surfactants exhibited poor stability, characterized by separation and sedimentation. Gum arabic-based nanofluids surfactant demonstrated immediate separation after ultrasonication, though this behavior was independ of time. However, at a higher gum arabic concentration (1:2), improved stability was observed compared to lower concentrations. Nanofluids stabilized with citric acid at relatively low concentrations showed excellent stability.

The most stable $NiFe_2O_4$/water nanofluid was achieved with a $NiFe_2O_4$-to-citric acid ratio of 1:0.25, yielding a zeta potential of -30 mV and an average thermal conductivity of 0.585 ± 0.007 W/m·K. When compared to the base fluids, the maximum enhancement in thermal conductivity of the obtained nanofluid was approximately 6%. Furthermore, the thermal conductivity enhancement is expected to be more pronounced with increasing nanoparticle concentration and in the presence of an applied external magnetic field, which is crucial for the optimizing the performance of the nanofluid.

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