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Investigation and optimization of effective factors on the thermal conductivity and stability of water-based CuO nanofluid

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***Abstract*— Nowadays it was proved that changing the thermal characteristics of the fluids is the best solution to increase the rate of heat transfer. One way is the use of suspended solid particles in a liquid due to the high thermal conductivity coefficient of them compared to liquids. This issue led to introduced a new concept that was called nanofluids. In this study, the water-based nanofluids with CuO nanoparticles are prepared by dispersing nanoparticles using an ultrasonic equipment. We prepared nanofluids of different weight concentrations with the addition of different particle sizes and various concentrations of surfactant. Thermal conductivities of these nanofluids were measured using the transient Hot-wire method. The main aim of this paper is to discuss the effects of the most important factors on nanofluids to provide the optimal conditions to achieve the highest thermal conductivity and stability of nanofluids. So, with the selection of different concentrations of water-Copper(II) oxide nanofluids as well as changes of pH values, it was determined that an aqueous nanofluid containing 1wt% CuO nanoparticles, pH=4 with 20 nm particle diameter, exhibited a thermal conductivity 47% greater than that of water. This nanofluid was chosen as the optimal nanofluid due to the thermal characteristics and desirable stability.**

Keywords-Nanofluids; Stability; thermal conductivity; CuO; effective factors; optimized properties

I. INTRODUCTION

One of the most significant scientific challenges in the industrial area is heat transfer enhancement. Because the public fluids used in the industry, such as water, ethylene glycol, pumping oil, etc. have not shown sufficient capability for heat transfer applications due to their poor thermal performance [1]. So far, various methods has been suggested to increase the heat transfer rate in fluids by authors. Including the use of extended surfaces (fins), reducing the heat resistance, increasing of the heat transfer coefficients (thermal conductivity or convection), etc. [2]. The researchers believe that changing the thermal characteristics of the fluids is the best solution to increase the rate of heat transfer. One way is the use of suspended solid particles in a liquid due to the high thermal conductivity coefficient of them compared to liquids. This issue led to introduced a new concept that was called nanofluids. Choi introduced the novel concept of nanofluids by applying the unique properties of nanofluids at the annual Mechanical Engineering meeting of American Society at 1995 [3]. Nanofluids, defined as suspended nanoparticles with the size of 1 to 100 nm inside fluids.

In the recent years, nanofluids have attracted great interest because of their higher values of thermal conductivity than those of commonly-used single-phase fluids. Nanoparticles have unique properties, such as large surface area to volume ratio, dimension-dependent physical properties, and lower kinetic energy, which can be exploited by the nanofluids [4]. Many mechanisms have been proposed to describe the anomalous thermal conductivity increase in nanofluids. Among these models the famous Hamilton-Crosser (HC) model was based on the Maxwell's model, and both of them only take into consideration of the volume fraction and the geometry of particles. HC model gives a good description of systems with micrometer or larger-size particles, but fails to predict the measured thermal conductivity of nanofluids [5]. Therefore, the thermal characteristics of nanofluids Should be study Using experimental methods. Eastman et al. measured the thermal conductivity of nanofluids. They found that an aqueous nanofluid containing 5% (v/v) CuO nanoparticles exhibited a thermal conductivity 60% greater than that of water. Additionally, they reported a 40% greater thermal conductivity compared to water for an aqueous nanofluid containing 5 % volume fraction of Al_2O_3

nanoparticles [6]. Various studies of researchers suggest that the most important factors affecting the thermal conductivity of nanofluids are temperature, concentration, pH, particle size, mixing method of the nanoparticles in fluids, etc. [7-9]. Although the researchers have similar opinion about the impact of these factors on thermal conductivity enhancement of nanofluids, but there are different conclusions in their reports about the thermal behavior prediction of the nanofluids affected by these factors. e.g. Angue Mintsa et al [10] reported that Increasing the temperature causes enhancement on thermal conductivity of nanofluids. Whereas Masuda et al [11] have reported that with increasing in temperature, the thermal conductivity of nanofluids decreased. Paul and Manna [12] also collected results presented by various researchers on the impact of temperature on the thermal conductivity of nanofluids. They concluded that temperature has the greatest impact on enhancing the thermal conductivity of nanofluids. But there is the most disagreement on the impact of nanoparticle size. Michael P. Beck et al [13] resulted that the thermal conductivity generally decreases with decreasing particle size below a certain particle diameter (~50 nm). They also reported the thermal conductivity of nanofluids containing larger particles appears nearly constant with particle diameter. Additionally, the thermal conductivity decreased as the particle size decreased for the nanofluids consisting of alumina in ethylene glycol. On the other hand, Maddah et al [14] and Tun-Ping Teng et al [15] observed an increase in the thermal conductivity of the nanofluids as the particle diameter decreased. Li et al. [16] recommended simultaneous control of both the pH and chemical surfactant to improve the thermal conductivity of Cu/H_2O nanofluids for practical applications. They reported a maximum thermal conductivity enhancement of 10.7% at 0.10% weight concentration. The effective thermal conductivities of Al_2O_3/water nanofluids with low volume concentrations from 0.01% to 0.3% were measured at 21℃ by Lee et al. [17]. They observed a maximum enhancement of 1.44% at a volume concentration of 0.3%.

There seems to be a consensus in various studies of researchers on the impact of concentration on thermal conductivity of nanofluids. Considering mentioned description, it is clear that beside the study on the impact of various factors experimentally, it should be provide an appropriate mechanism to predict the thermal characteristics of nanofluids affected by various factors. Hence, several new concepts and mechanisms have been proposed in the last decade to account for this anomalously enhanced thermal conductivity. Some of these proposed mechanisms include nanolayer, aggregation, percolation like behavior, interfacial thermal resistance, Brownian motion of nanoparticles, nanoconvection and surface charge mode. Hot debates are ongoing in the nanofluids community on the validity of proposed mechanisms and models of heat conduction in nanofluids. A number of mechanisms and models of enhanced conductivity have been proposed, but none has gained universal support [18]. Preparing a homogeneous suspension is still a technical challenge due to strong van der Waals interactions between nanoparticles always favoring the formation of aggregates. To obtain stable nanofluids, some methods are recommended, such as physical or chemical treatment [1]. Addition of surfactant can improve the stability of nanoparticles in aqueous suspensions. The reason is that the hydrophobic surfaces of nanoparticles/ nanotubes are modified to become hydrophilic and vice versa for non-aqueous liquids [1].

Research also showed that the thermal conductivity of nanofluids is time dependence immediately after dispersed by ultrasonication, then time independence for the longer time [19]. It has been verified experimentally that surfactant is effective in dispersing nanoparticles in base fluids and weakening the agglomeration behavior in nano-suspensions [20-22]. Yu et al. [20] prepared stable ethylene glycol-based copper nanofluids with the addition of polyvinyl pyrrolidone (PVP) as surfactants. Results showed that the addition of PVP significantly improved the stability of copper nanofluids, yet had negative effects on the thermal conductivity. Yang et al. [21] reported that the presence of surfactants (PEG, PAA) could improve the stability and thermal conductivity of nanofluids, but surfactants in low concentrations had smaller influence than other influence factors such as particles and temperature on the thermal conductivity. Zhou et al. [22] experimentally investigated the thermal conductivity of several common surfactant (SDS, SDBS, CTAB, PVP) solutions, concluding that the thermal conductivities of surfactant solutions reach a stable ratio after a certain concentration, and the thermal conductivity ratios of ionic surfactant solutions are higher than those of non-ionic surfactant. Addition to using the surfactants to improve the stability of nanofluids, it is very important the pH adjustment of nanofluids. Because The stability of an aqueous solution nanofluid directly links to its electrokinetic properties. Through a high surface charge density, strong repulsive forces can stabilize a well-dispersed suspension. The isoelectric point (IEP) is the concentration of potential controlling ions at which the zeta potential is zero. Thus, at the IEP, the surface charge density equals the charge density, which is the start point of the diffuse layer. Therefore, the charge density in this layer is zero. As the pH of the solution departs from the IEP of particles the colloidal particles get more stable and ultimately modify the thermal conductivity of the fluid. The surface charge state is a basic feature which is mainly responsible for increasing thermal conductivity of the nanofluids [23,24].

As stated above, there were already many works studying the influence of factors such as temperature, particle size, pH control, surfactant etc. on the thermal conductivity and stability of nanofluids, individually. Hence with the objective to contribute to the expanding nanofluid properties database, experimental investigations of thermal conductivity and stability measurements on CuO/water nanofluid are made and the results are presented in this paper. In this work, effects of Sodium dodecyl sulfate (SDS) and Sorbitan monolaurate (Span60) as surfactant materials on the stability and thermal conductivity were analyzed. We prepared CuO/de-ionized water nanofluids of different weight concentrations with the addition of different particle sizes and various concentrations of surfactant. Thermal conductivities of these nanofluids were measured using the transient Hot-wire method. The main aim of this paper is to discuss the effects of the most important factors on the thermal conductivity and stability of nanofluids to provide the optimal conditions to achieve the highest thermal conductivity and stability of nanofluids.

II. MATERIALS AND METHODS

A. Materials

Hydrophilic spherical nanoparticles of CuO (supplied by Degussa, Germany) with two different nanoparticles size were purchased in the forms of dry powder. Physical properties of these nanoparticles are given in Table 1. The mean grain size of the these particles are about 20 nm and 50 nm. De-ionized (DI) water (99% purity) was used as base fluid. pH adjustment of nanofluids was achieved through analytical grade 0.1 M NaOH and HCl. It was also used Sodium dodecyl sulfate (SDS) and Sorbitan monolaurate (Span60) (they both supplied by Sigma-Aldrich, Germany) as surfactant materials. In order to stabilize nanofluids it was used ultrasonic bath (SW 1 H model made by Sonoswiss) and Magnetic stirrer (VS-130SH). To measure thermal conductivity was used Transient Hot-Wire method. This device was constructed based on the copper wire as a thin wire by authors. The stability of nanofluids were determined using Ultraviolet-Visible spectrophotometry (UV-Vis) (model UV-2502 Labomed, Inc.).

B. Preparation of nanofluids

Nanofluids with different weight concentrations were prepared by dispersing specified amounts of CuO nanoparticles in DI water. Thus samples were prepare with 0.5, 1, 1.5 and 2wt% copper (II) oxide. Then they were firstly placed in the magnetic stirrer for 30 min then they were exposed in the ultrasonic bath for 2 h to break down the agglomerations. To investigate the effect of temperature on the thermal conductivity, the samples were measured at a specific temperature range (5 to 50°C). For stability comparison, suspensions with any surfactants were prepared in the same way. Hence surfactant solutions were prepared and transferred into an ultrasonic bath according to the critical micelle concentration (CMC). CuO powder was added to the surfactant solutions gradually in the ultrasonic bath at 25°C.

III. RESULTS AND DISCUSSION

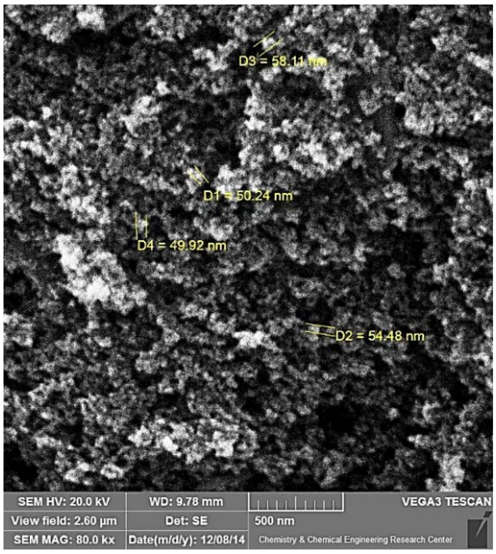
Fig. 1a and b show the SEM images for CuO nanoparticles. The powder sizes of CuO nanoparticles are 45–50 nm and 15–20 nm, respectively. These figures show that the shape of the nanoparticles are rather spherical. In this study, the effective thermal conductivity of DI water based nnanofluids with nanoparticles weight fraction ranging from 0.5 to 2wt% are measured at the temperature ranging from 5 to 50°C.

Table 1. Physical properties of CuO nanoparticles

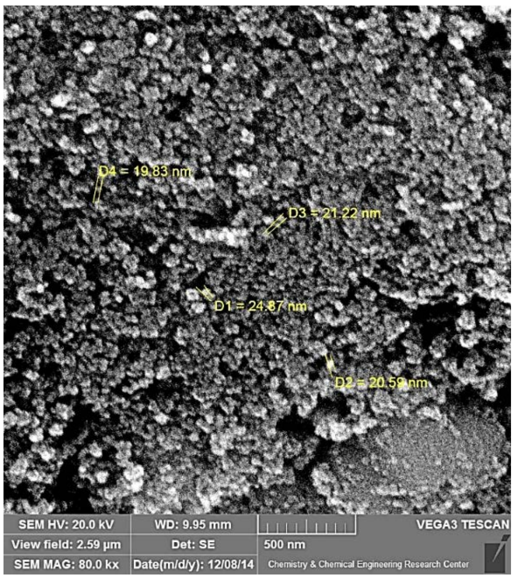
CuO	Ca	Fe	Cr	Na	Mn
99%	<25ppm	<80ppm	<4ppm	<60ppm	<2ppm

Fig. 2 shows the thermal conductivity enhancement of DI water-based 1wt% CuO (20 nm) nanofluid as a function of temperature. As seen, thermal conductivity increases with temperature due to the number of collisions between nanoparticles and molecules base fluid increase which it makes the transfer of thermal energy between the particles of medium can be occur more quickly than lower temperatures. Also in order to investigate the models proposed by researchers such as Bruggeman model (Eq. 1) the results were compared with them.

$$k_{eff} = \left\{ \frac{1}{4}\left[(3\phi-1)\frac{k_p}{k_f} + (2-3\phi) \right] + \frac{1}{4}\sqrt{(3\phi-1)^2\left(\frac{k_p}{k_f}\right)^2 + (2-3\phi)^2 + 2\left(2+9\phi-9\phi^2\right)\left(\frac{k_p}{k_f}\right)} \right\} k_f \quad (1)$$



(a)



(b)

Figure 1. SEM photograph of CuO particles a) 50 nm, b)20 nm.

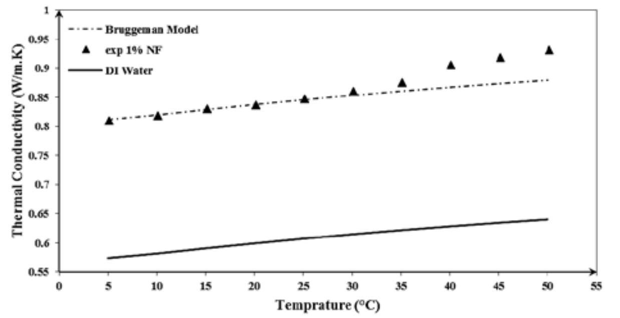


Figure 2. thermal conductivity enhancement of DI water-based 1wt% CuO (20 nm) nanofluid as a function of temperature.

As is evident from Fig. 3a and b, the general trend shows a gradual increase in the thermal conductivity ratio of nanofluids with an increase in nanoparticle loading concentration for 20 nm and 50 nm particle size respectively. it is observed that the thermal conductivity increases with the increase in weight fraction,which is almost linear. A small variation is observed in measured thermal conductivity compared to the predicted values obtained from the Bruggeman and HC models (Eq. 2). Thus, the proposed models are capable to predicte the thermal properties of nanofluids with different concentrations whereas it has not been considered the effects of temperature, pH, particle size etc. on them.

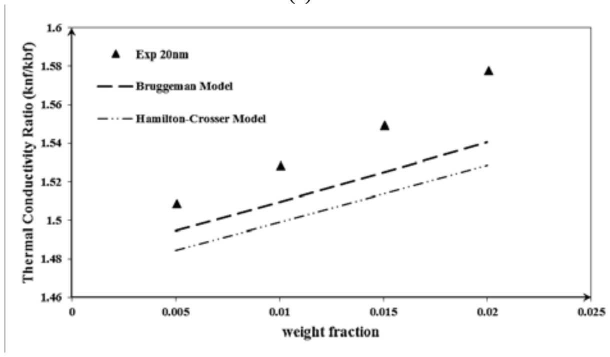
$$k_{nf} = \left[1+3\beta\phi+\phi^2\left(3\beta^2+\frac{3\beta^3}{4}+\frac{9\beta^3}{16}\frac{\chi+2}{2\chi+3}+\frac{3\beta^4}{26}+...\right)\right]k_{bf} \quad (2)$$

$$\chi=\frac{k_p}{k_{bf}}, \beta=\frac{k_p-k_{bf}}{k_p+2k_{bf}}$$

For analyzing the effect of particle size on the thermal conductivity of nanofluids, results have been reported in Fig. 4. The thermal conductivity ratio as a function of weight concentration for different particle sizes are shown in Fig. 3. It can be easily seen from the figures that for the same type of particle and base fluid medium, the thermal conductivity ratio for a smaller sized particle is much higher than that for a larger sized particle. This observation is valid for all types of particles and base fluid mediums. This is more clearly demonstrated in Fig. 4, which graphically represents the thermal conductivity ratio as a function of particle size for 20 nm and 50 nm nanoparticles. The general trend is a decrease in thermal conductivity ratio with an increase in particle size. But as previously mentioned, there is diverse reports about the effect of particle size on thermal conductivity. As stated Beck et al. [13] report that for magnetite–water nanofluids, the thermal conductivity ratio increases considerably with an increase in particle size. In most of these cases, particle agglomeration being a key factor that cannot be determined from experiment or theory may contribute largely to the ambiguity of results produced.

At the isoelectric point of the solid, its zeta potential is zero. This is found by adjusting the pH to the appropriate value. In order to analysis the effect of isoelectric point on the thermal conductivity and stability of CuO/de-ionized water nanofluids with different particle sizes, suspensions with different concentrations CuO of 20 nm and 50 nm particle diameter were prepared by adjusting of pH value in range of 3 to 11.6. Results of thermal conductivity measurement for 20 nm and 50 nm particle diameter are shown in Figs. 5 a and b respectively.

(a)



(b)

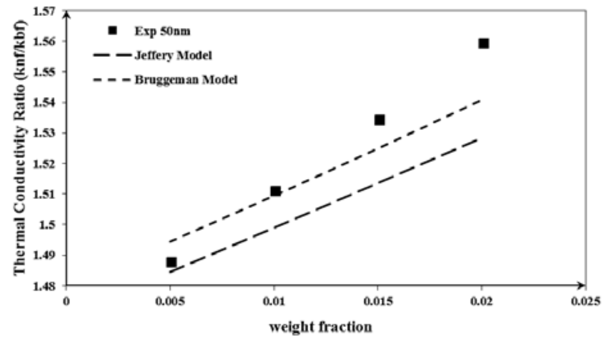


Figure 3. Thermal conductivity of CuO/water nanofluids at different volume fractions and at 25 °C. a)20 nm, b)50 nm

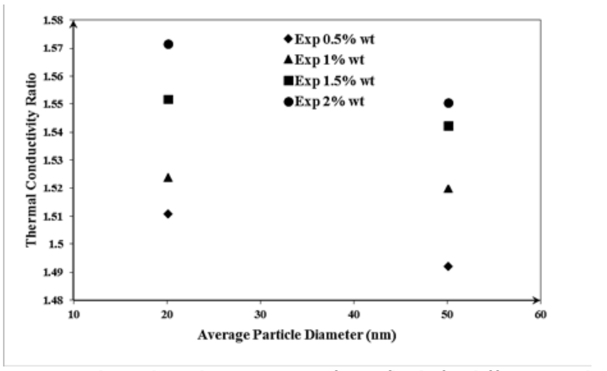


Figure 4. Thermal conductivity ratio of nanofluids for different sized particles as a function of concentration at 20°C.

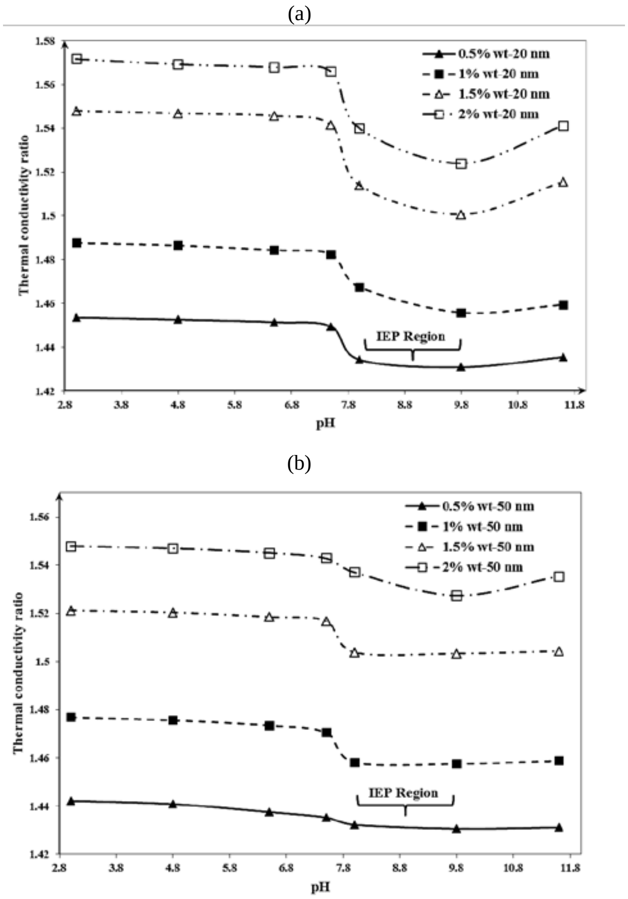


Figure 5. Thermal conductivity measurement for 20 nm (a) and 50 nm (b) particle diameter as function of pH

In order to analysis the effect of surfactants on the thermal conductivity of CuO/de-ionized water nanofluids, suspensions with 1 and 2wt% CuO of 20 nm particle diameter were prepared by adding different concentrations of SDS and Span60. Surfactant has negative effects on the heat conduction of base fluid. Due to the longer alkyl chain length which causes reduction in the thermal conductivity of nanofluid. But Span60 is a cationic surfactant and has low HLB (Hydrophile-Lipophile Balance) 4.7 value. The HLB of an surfactant is an expression of its Hydrophile-Lipophile Balance, i.e. the balance of the size and strength of the hydrophilic (water-loving or polar) and the lipophilic (oil loving or non-polar) groups of the emulsifier. An emulsifier that is lipophilic in character is assigned a low HLB number (below 9.0) that they are not suitable for aqueous solutions. Thus by adding the Span60 with any concentration to the suspensions makes it two-phase quickly. Therefore Span60 did not satisfy the increased stability of nanofluids, contrast to the SDS. Results of thermal conductivity ratio are shown in Fig. 6 for SDS surfactant. As presented in the figure, the thermal conductivity ratio of 20 nm CuO/de-ionized water nanofluids change with different concentrations of SDS. The highest thermal conductivity ratios of 20 nm CuO/de-ionized water nanofluids occur when the concentration of SDS increases to 1.0 wt%, that it is the critical micelle concentration (CMC) for SDS surfactant.

Fig. 7 shows the weight fraction change of the nanofluids with the passage of time. The weight concentrations of DI water-based CuO nanofluids vary by particle aggregation and sedimentation. After 30 days days of the nanofluid preparation, the weight fraction of the water-based CuO nanofluids decreased for any concentrations of copper(II) oxide. However, the results are different as compared to when the SDS has been used. As shown in Fig. 8 with addition of surfactant the stability of nanofluid rised so that stability is desirable for low concentrations. Fig. 8 represents the weight fraction change of 1wt% nanofluid with 20 nm particle diameter as function of pH value. The results confirm that the remoteness of the pH value from isoelectric point makes the stability further enhanced.

Fig. 9 shows the thermal conductivity variation of the nanofluids with the passage of time. As mentioned earlier, surfactant is a negative factor for thermal conductivity enhancement due to the alkyl chain length, which decreases the thermal conductivity of nanofluids.

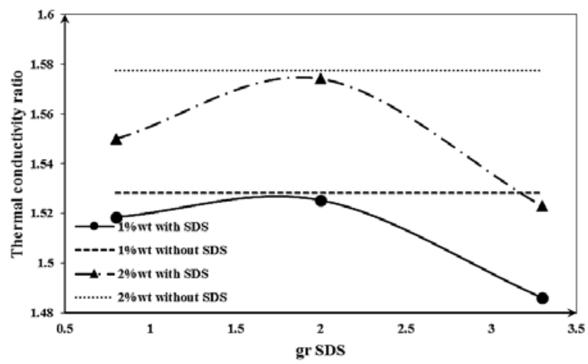


Figure 6. Influence of SDS concentration on the thermal conductivity of CuO/deionized water nanofluids at room temperature.

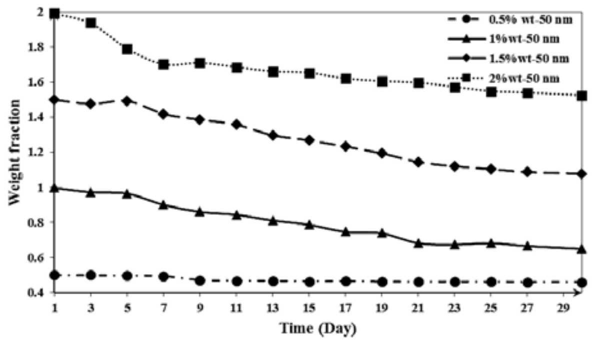


Figure 7. Weight concentration change of various nanofluids with the elapse of time

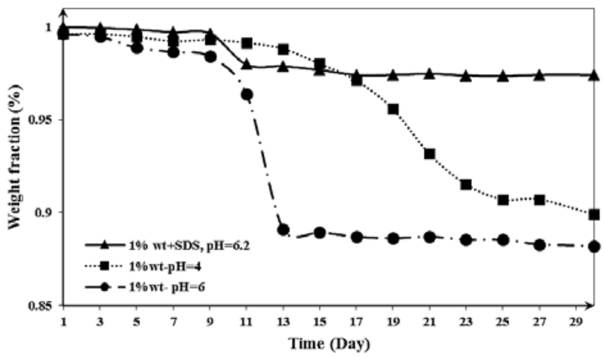


Figure 8. Weight concentration change of 1wt%+SDS nanofluid as function of pH value with the elapse of time

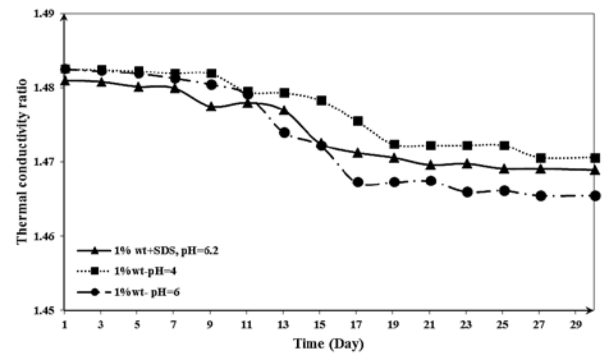


Figure 9. Thermal conductivity change of 1wt%+SDS nanofluid as function of pH value with the elapse of time

IV. CONCLUSIONS

A nanofluid is a particle suspended fluid, so there are particle–particle and particle–fluid interactions that can lead to the aggregation and sedimentation of particles. In this study, the thermal conductivity of DI water based CuO nanofluids with different particle sizes were measured by Transient hot wire method. The influence of the weight concentration, temperature, pH control, surfactant and the particle size variation on the thermal conductivity and stability of nanofluids were studied. So, with the selection of different concentrations of water-copper(II) oxide nanofluids as well as changes of pH values, it was determined that the best nanofluids prepared with 1wt% (pH=4) and consisting of 20 nm nanoparticles. Therefore, it's appropriate for thermal conductivity enhancement of base fluid and has a good stability with time. The results showed an aqueous nanofluid containing 1wt% CuO nanoparticles, pH=4 with 20 nm particle diameter, exhibited a thermal conductivity 47% greater than that of water. This nanofluid was chosen as the optimal nanofluid due to the thermal characteristics and desirable stability. While the features such as high thermal conductivity and stability, are not available for other nanofluids simultaneously.

the particle size measurement results demonstrate that the particles in a nanofluid undergo sedimentation, aggregation+sedimentation and aggregation in time order.

The pH control, which has an important role in stability control, places the IEP of the suspension, far from the PZC in order to avoid coagulation and instability. It should be taking into account that acidic or alkaline pH is corrosive to metals. Therefore, it can lead to damage to the piping and instrumentation in long term applications. Therefore, the value of 4 was chosen as the optimum pH value.

Surfactant selection in nanofluid preparation has an important role in improving stability of nanofluids. Temperature is considered as a restricted factor in case of nanofluid application for exploiting at the high temperatures. Likewise, the optimum percentage of surfactant should be considered as a factor in stable nanofluid preparation as well. Thus it is necessary to identify the CMC values of any surfactant which is used.

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