Stability Suspension of Al_2O_3 and ZnO in Ethanol-Water Mixture with Dependence of Sonication Time

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Abstract - **Nanofluids are being studied for many heat transfer application in the enhancement of thermal conductivity. The nanoparticles dispersed in the conventional base fluid tend to settle with time and it is one of the challenges for its stability as a heat transfer fluid. Sonication is one of the methods that have been used to increase the dispersion of nanoparticles and increase the nanofluids stability. In this study, stability suspension of alumina (Al_2O_3) and zinc oxide (ZnO) nanoparticles, of 50 nm and 40 nm respectively, in ethanol-water mixture are observed with dependence of sonication time of 10 minutes, 20 minutes and 30 minutes of probe sonication. Sedimentation method is used to study its stability based on sonication time and the concentration of both nanoparticles of 0.5 wt %, 1 wt % and 3 wt % in the ethanol-water mixture of 0%, 10%, 30%, 50%, 70%, 90% and 100% concentration. Sedimentation test of each sample has been critically checked and recorded. As per result, after 24 hours, nanofluids that has been sonicated for 30 minutes shows higher sedimentation ratio and better stability than samples with 20 minutes sonication. 10 minutes sonicated samples sediment immediately after sonication. At instantaneous time, 50 wt % of ethanol-water mixture is most stable than other range of concentration, and as the nanoparticles concentration increased, the suspension become more stable. However, in a longer period, different ethanol-water concentration exhibit most likely trend by means of stability. By comparison, alumina samples are more stable than ZnO samples. Therefore, the stability suspension of alumina and ZnO without surfactant is affected by sonication time, and the concentration of the nanofluids.**

Keywords – Alumina (Al2O3) and Zinc Oxide (ZnO; Ethanol-Water Mixture; Stability Suspension; Sedimentation Method; Sonication

I. INTRODUCTION

Nanoparticle is the particle of size 1-100nm and it may have properties that are different from its parent material [1]. It has been introduced in nanotechnology to enhance heat transfer application compared to conventional heat transfer fluid. Nanoparticles are dispersed in base fluids to form nanofluids that have the ability to enhance heat transfer application. Heat transfer enhancement can be achieved by adding high thermal conductivity nanoparticle such as alumina (Al_2O_3) and zinc oxide (ZnO) to a lower thermal conductivity base fluid. In developing nanofluids for a wide range of heat transfer application, the understanding of fundamentals heat transfer is important [2]. However the challenges in further development of nanofluids are there as the nanoparticles tend to sediment and to settle down after some period due to gravity forces. This is because small particles possess high surface energies that gives large amount of ionisable sites, thus causes higher tendency to build agglomerates among them [3]. Agglomeration not only results in settlement but also in decreasing overall thermal conductivity of nanofluids [4]. Therefore, stability suspension is one of the important factors to take into account in order to produce a better approach of heat transfer enhancement using nanofluids.

Current researches have shown that stability suspension is one of the dilemmas that may affect the properties of nanofluids for heat transfer enhancement technology. Sonication is clearly helpful to increase the stability suspensions, but vary according to type of nanoparticles, type of base fluids, concentration, viscosity, pH of the nanofluids and the sonication time itself. Moreover, the stability also tested using sonication with and without surfactant. Henceforth, stability suspension of Al_2O_3 and ZnO in ethanol water mixture in dependent of sonication time, without surfactant is anticipated in this study.

The focus of the study is to observe the effect of sonication time on nanofluids stability suspension. The sedimentation trend of nanoparticles at different concentration with sonication is to be studied. Moreover, the stability suspension of Al_2O_3 and ZnO in ethanol-water mixture with sonication is compared.

The aim of this study is to improve the sedimentation study for stability suspension of nanofluids. The study about sedimentation study on nanofluids is not new since there are many researches have been done to validate the agglomerate-free stable suspension as it is the most important criterion of nanofluid [5]. Withrana, *et al.* [6] claimed that Al_2O_3 in general is more stable in all type of base fluids in their experiments using propylene glycol, ethylene glycol, and mixtures of both in water [7]. Ethanol-water mixture with different concentration as base fluids is used, and the sedimentation of Al_2O_3 and ZnO nanoparticles are recorded.

II. LITERATURE REVIEW

A. Application of nanofluids

Nanoparticles are made of tiny particles of measurement 1-100nm and able to interact at molecular level and exhibit different properties from their parent materials [1]. Nano particles has higher thermal conductivity and they are added to base fluids to form nanofluids; a colloidal suspension. By suspending nanoparticles in these base fluids, they are expected to enhance heat transfer capability of the conventional heat transfer fluid, due to their higher thermal conductivity. The physical size of nanoparticles gives them ability to flow easily in micro-channel without clogging, and with higher heat transfer efficiency, the size of heat transfer system can be reduced [4]. It can be applied in many heat transfer application including cooling processes, manufacturing, medical treatment and electrical application [8].

Nanofluids can be prepared using two methods; one-step method and two-step methods. One-step method may reduce the agglomeration of the nanoparticles and it is a continuous process of making and dispersing the particles in the fluid. Few steps are skipped in this method, hence agglomeration of nanoparticles can be reduced and the stability of the nanofluids is better. However, this method is will consume higher cost in large scale synthesis [9]. In two-step method that is also known as dispersion method, nanoparticles are in the form of dry powder and it will be dispersed into base fluid. This step may cause agglomeration of the nanoparticles but can be reduced by ultrasonication [8]. However, this method is the most economic method to produce nanoparticles in large scale.

Alumina and ZnO in Ethanol-Water Mixtures

Ethanol-water has shown its good agreement with surface tension, better than water itself thus it may reduce the agglomeration of nanoparticles in the solution [10]. It is also proved that the combination of ethanol and water gives higher viscosity and it reduces any vortex formation and opposes mixing of other particles [11].

Alumina and ZnO are among the nanoparticles that are widely tested in stability suspension of nanofluids studies. Alumina has been claimed in many studies that it gives a better stability in all type of base fluids such as water, ethylene glycol, and propylene glycol [6] but the stability may be affected by the volume fraction of alumina in water [5] and it depends on the sonication time introduced to the nanofluids [4]. On the other hand, ZnO is claimed to be stable only in propylene glycol base fluids after 4 hours of sonication [6], however, it is suggested that ZnO nanoparticles to be modified by polymethacrylic acid (PMAA), a type of surface modification technique, in aqueous system [8]. In addition, horn sonication is found to be more effective on ZnO [13].

B. Stability suspension of nanofluids

Stability suspension is one of the important criteria of nanofluids in order to perform well in heat transfer enhancement for further possible application. A stable nano-suspension is the one with agglomerate-free and stable in longer period [5] and its properties only valid as long as it is stable [3]. Due to its high-surface activity, nanoparticles tend to agglomerate with the time elapsed and this agglomeration can reduce the thermal conductivity of the nanofluids, and the settlement and clogging in micro channel may occur [4]. This phenomenon of agglomeration is characterized by the formation of particle clusters as the particles are in contact with each other and cohesion takes place [14]. Factors that influenced the stability of suspensions including its concentration, dispersant, viscosity of base fluid and pH value [15]. In addition, the variety, diameter, density of nanoparticles and ultrasonic vibration can also influence the stability of the nanofluids. To improve the stability of nanofluids, general methods such as change of pH value, addition of surfactant, surface modification and ultrasonication can be applied. Besides, agglomeration can be avoided by oven drying the nanoparticles before dispersing them into base fluid as it will remove the moisture trapped inside the nanoparticles [16].

Previous study has shown positive results in using sonication to test the stability suspension of nanofluids. Ultrasonication gives better dispersion result [1]. However, too long time of high-energy sonication may introduce defects to the particle size distribution [23]. As define by Stokes' Law [25], sedimentation of nanoparticles can be reduced by reducing its size or increasing the viscosity of the base fluids, or minimize the density between the nanoparticles and the base fluids [3, 26].

Sedimentation study

Sedimentation study is the most conventional method to evaluate the stability suspension of nanofluids. Using special apparatus and various concentrations of nanofluids, sedimentation data can be obtained [8]. In sedimentation study, ultrasonication can be used to avoid agglomeration of nanoparticles after adding it into base fluids using two-step method, and the sedimentation height in test tube is measured in dependence of time. The more stable the nanofluids, the longer it takes to settle down, and more Ultrasonication time gives better dispersion result [1]. The sedimentation photograph is taken to observe the stability of nanofluids [4].

There are many types of ultrasonication equipment can be used to disperse the nanofluids. Two of the common types are bath sonicator and probe sonicator. It is proven that probe sonicator will give more efficient dispersion than bath sonicator [27].

III. METHODOLOGY

A. Materials

The alumina and ZnO powder of the size about 50nm and 40 nm respectively, are used in this study. Ethanol of 99.7% concentration is diluted to different concentration of 10%, 30%, 50%, 70%, and 90% using deionized water.

The nanofluids are then prepared using two-step methods, using different parameters as shown in Table 1. All nanofluids are prepared without using any surfactants or surface stabilization additives.

Table 1: List of parameters used in the experiment

Variables	Parameters
Controlled Variables	No surfactant or additives Size of nanoparticles Sonication settings: 80 W, 90% pulser, 20kHz
Independent Variable	Sonication time: 10 minutes, 20 minutes, 30 minutes Concentration of ethanol-water: 0%, 10%, 30%, 50%,70%, 90% 100% Weight Fraction of nanoparticles: 0.5%,1%, 3%
Dependent Variables	Sedimentation height of nanofluids

B. Experimental Equipment

Sedimentation apparatus that are going to be used in this experiment includes flat bottom test tubes with caps, ruler, dark backdrop, and timer. To vary the concentration of the nanofluids, standard laboratory apparatus such as conical flask and volumetric flask are to be used. Digital camera is needed as well to get the photograph for the observation of nanofluids sedimentation.

Figure 1: Biology.Inc Model 150 VT Ultrasonic Homogenizer that is used for sonication

The sonication will be done using Probe Sonicator (Biology,Inc Model 150 VT Ultrasonic Homogenizer) as it is effective to agitate nanomaterials. The sonication will be done for 10 minutes, 20 minutes and 30 minutes for every sample to study the sedimentation of nanoparticles in the base fluids with dependence of sonication time. In addition, before the samples are prepared, drying oven is used to remove moisture from nanoparticles to avoid agglomeration due to ionisable sites. Bulk drying methods like oven drying is used for drying nanoparticles at the laboratory scale [16].

IV. RESULT AND DISCUSSION

Experiment was conducted by manipulating the concentration of ethanol-water mixture, and the concentration of the nanoparticles. The sonication time is varied to 10 minute, 20 minutes and 30 minutes, makes the total is 126 samples.

From the sedimentation observation, the picture of all sonicated samples are recorded and calibrated technically. This is to ensure the precision and the accuracy of the results. The pictures were captured using an 8 Megapixels camera in the scale of 1 cm. Figure 2 shows on how the measurement is taken.

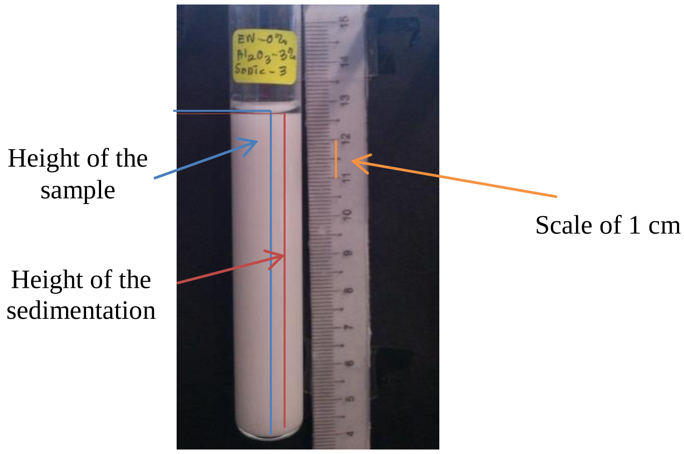

Figure 2: Observation after 15 minutes for sample of 0 wt% of ethanol-water mixture with 3 wt% of alumina after 30 minutes of Probe Ultrasonication

Sedimentation ratio is obtained by:

$$\text{Sedimentation ratio} = \frac{actual\ height\ of\ sedimentation}{actual\ height\ of\ sample}$$

Where, the actual height is obtained after the technical calibration with the 1 cm scale.

A. Effect of Sonication Time

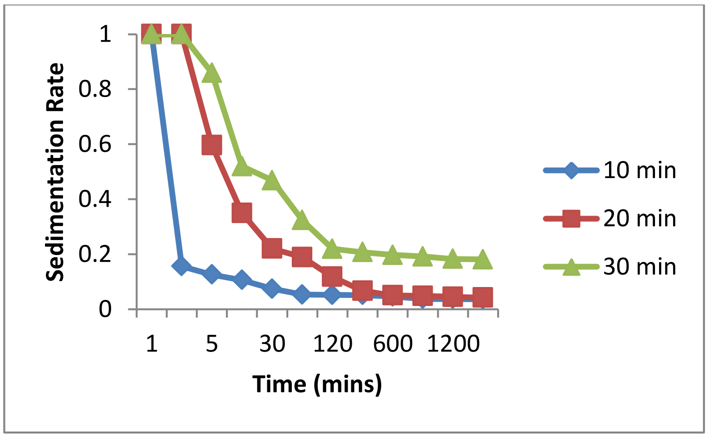

Figure 3: Sedimentation ratio of 0.5 wt % of Alumina in 0 wt % of ethanol-water mixture after 24 hours

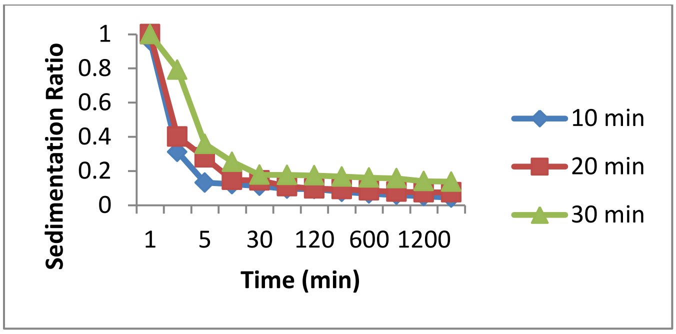

Figure 4: Sedimentation of 0.5 wt % of ZnO in 0 wt % of ethanol-water mixture after 24 hours

Figure 3 and 4 show a physical explanation of sedimentation test of alumina and ZnO for different time of sonication introduced. In Figure 10, the sedimentation ratio of alumina samples that were sonicated for 10 minutes immediately sediment after 2 minutes and the rate of sedimentation is high in the first 5 minutes of observation. Being more stable, rate of sedimentation of the samples that were sonicated for 20 minutes getting slower only after 15 minutes. For 30 minutes sonicated samples, the sedimentation rate is clearly slower than the other two sets of samples. ZnO samples show most likely trend as alumina. Thus, it is clearly shown that with higher sonication time introduced to the samples, the stability of the suspensions without using surfactants can be ensured.

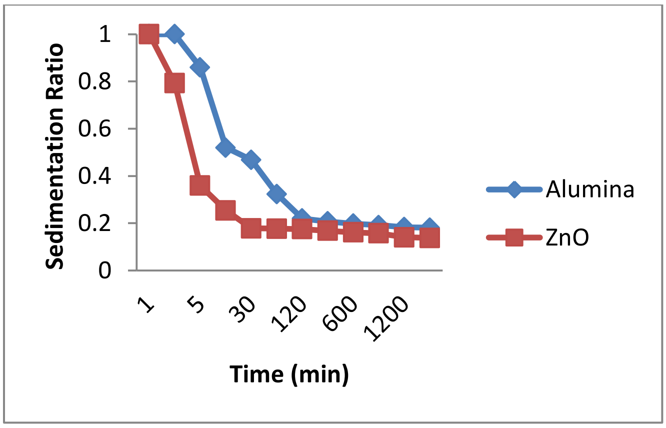

Figure 5: Sedimentation ratio of 0.5 wt % nanoparticles in 0 wt % ethanol-water mixture with 30 minutes of probe sonication, after 24 hours

In comparison, from Figure 5, alumina shows a better stability than ZnO. Rate of sedimentation is higher for ZnO since the sedimentation ratio drops after 30 minutes of sedimentation. It proves that alumina is almost stable in any base fluids [6].

B. Effect of Ethanol Water Concentration

The concentration of the base fluids is also tested at 0%, 10%, 30%, 50%, 70%, 90% and 100% of ethanol in water. Hence, the sedimentation ratio in this range of concentration is observed.

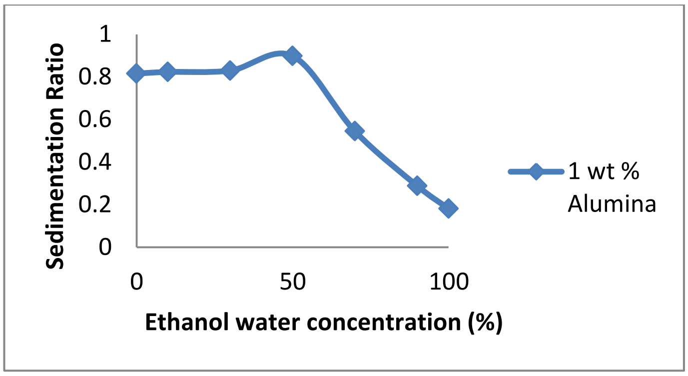

Figure 6: Sedimentation ratio of 1 wt % Alumina at different concentration of ethanol-water mixture with 10 minutes of probe sonication at instantaneous rate of 15 minutes of observation

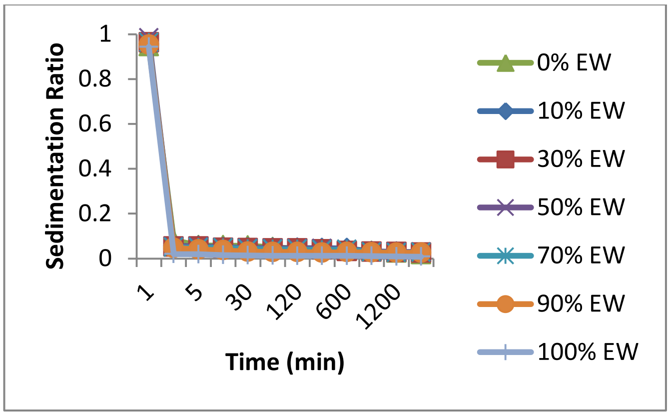

Figure 7: Sedimentation ratio of 0.5 wt % ZnO with 10 minutes of probe sonication at different concentration of ethanol-water mixture after 24 hours

In Figure 6, the sedimentation ratio at instantaneous rate of 15 minutes is taken and analyzed. It can be seen that the sedimentation ratio of the suspension 1 wt % alumina is most stable in 50 wt % of ethanol-water mixture and reduces as the concentration of ethanol is higher. Based on Wensinck [12], the occurrence is due to the effective polarization of these two molecules. The agglomeration is hard to occur if the concentration is below the limit, however, the agglomeration will occur if the concentration is beyond the limit, hence, leading to sedimentation [14].

However, after 24 hours of sedimentation, it can be observed in Figure 7 that the sedimentation trend of the ZnO suspensions almost similar for all range of ethanol water mixture concentration. Hence, it can be said that the ethanol-water concentration might not give significant effect on the stability suspension of the nanoparticles after longer observation time.

C. Effect of Nanoparticles Concentration

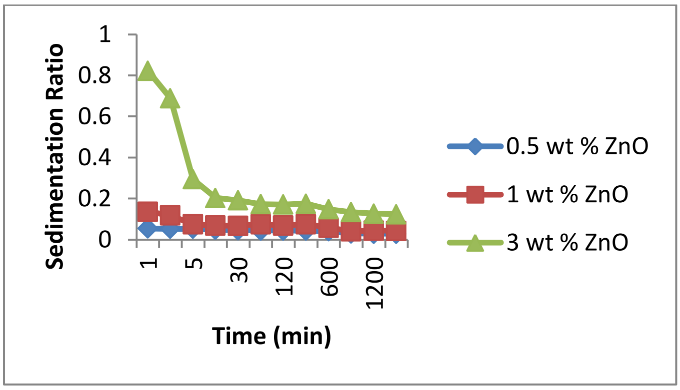

Figure 8: Sedimentation ratio of ZnO in 50 wt % ethanol-water mixture with 10 minutes of probe sonication after 24 hours

In Figure 8, sample of 0.5 wt % and 1 wt % of ZnO sediment immediately after 10 minutes of sonication, whereas, the sedimentation rate for 3 wt % of ZnO is slower than the other two. It shows that the suspension is more stable with higher concentration of nanoparticles.

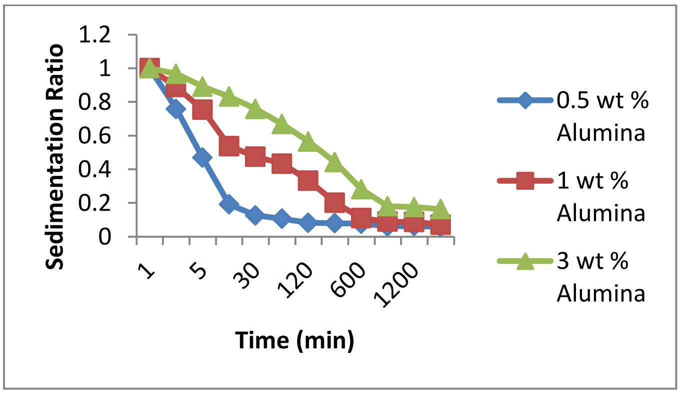

Figure 9: Sedimentation ratio of alumina in 50 wt % ethanol water mixture with 30 minutes of probe sonication after 24 hours

Figure 9 presents clearer trend of different concentration of alumina after 30 minutes of sonication. 3 wt % of alumina shows most stable suspension, and after 24 hours the sedimentation ratio is 0.16. On the other hand, 0.5 wt % of alumina shows least stable suspension and shows almost no rate at all after 10 hours and after 24 hours the sedimentation ratio decreased to 0.05.

D. Limitations

There are several limitations faced during the experiments. These limitations might affect the accuracy of experimental results.

a) Temperature control

During sonication, the power introduced to the solution with tends to heat up the solution, and the temperature of the solution will vary for every sample. According to literature [19], different surrounding temperature might affect the stability of nanofluids suspension. However, the equipment used does not have any features to control the temperature of the solution, hence; the effect of temperature is neglected.

b) Retention time for the solution

Before sonication, all samples are prepared at once. However, at sonication stage, the sample needs to be sonicated one by one, leaving the rest of the sample unattended. Due to this issue, some of the prepared solution may have agglomeration before sonication; make it more difficult to break the agglomerates during dispersion, hence, leads to poor stability. If the sample is prepared exactly before sonication, the results of the sedimentation study might be improved.

c) Power introduced

The maximum power for the probe sonicator used is 150 W. In this study, 80 W of power is used to ensure the solution is not overheated by the dispersion process. Nevertheless, testing the stability suspension of nanoparticles with dependence of sonication power of the particular equipment should be conducted to determine the optimum power of it before developing this study. Using optimum sonication power might improve the result of this study.

V. CONCLUSION

This study is to observe the stability suspension of alumina and ZnO in ethanol-water mixtures, in order to develop nanofluids characteristics for heat transfer enhancement using sedimentation method with dependence of time. Probe sonication is used to increase the dispersion of nanoparticles in the base fluid and to enhance the stability of the suspension.

As a conclusion, 30 minutes of probe sonication gives a better stability to the suspension compared to 10 minutes and 20 minutes dispersion. The higher the sonication time, the suspension becomes more stable. The suspension of alumina is more stable in a longer period than ZnO after 30 minutes of sonication. At instantaneous time, 50 wt % of ethanol water mixture shows highest sedimentation ratio, hence; more stable than other range of ethanol water concentration. However, after 24 hours of observation, different concentrations of ethanol-water mixture exhibit insignificant different on stability suspension. For different concentration of nanoparticles in the base fluid, as the concentration is increased, the stability of the suspension is enhanced.

Therefore, the quality of nanoparticles suspension without surfactants can be ensured with more sonication time introduced by reducing agglomeration problem.

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