Qualitative investigations on the stability of Al₂O₃-SiO₂ hybrid water-based nanofluids

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Abstract. Qualitative experiments were carried out to investigate the stability of Al₂O₃-SiO₂ hybrid water-based nanofluids. The main nanoparticles used in this work were Al₂O₃ and SiO₂ since these types of nanoparticles have been widely used and proposed in many applications and information on their hybrid dispersion stability characteristic are still elusive. The present work demonstrated the stability of Al₂O₃-SiO₂ hybrid nanofluids using various preparation techniques and successively elucidating the optimum preparation method for hybrid nanofluid. The hybrid water-based nanofluid were prepared by dispersing a 50:50 ratio of Al₂O₃-SiO₂ nanoparticles into distilled water to achieve total volume concentration of C=0.001, 0.01 and 0.1 vol%.

Successively, the hybrid nanofluids were prepared using magnetic stirrer and ultrasonic bath. The samples are categorised into five preparation method which are: (i) 1 hour sonication, (ii) 1 hour magnetic stirrer, (iii) 0.25 hour magnetic stirrer and 1 hour sonication, and (iv) 0.5 hour magnetic stirrer and 1 hour sonication and (v) without magnetic stirring and sonication. The height of sedimentation and colloidal stability of each preparation methods were observed in the duration of 2 weeks. In the present work, it was observed that the dispersion stability of hybrid nanofluids prepared by applying magnetic stirrer for 15 minutes with prior to 1 hour of sonication had demonstrated the highest stability indicated by the lowest height of sedimentation and high colloidal stability.

Keywords. Aluminium oxide; Silicon dioxide; Metal oxide hybrid nanofluids; Stability

1. Introduction

Nanoparticles can be defined as particles with less than 100nm size in at least 1 dimension. When nanoparticles are formed, the properties of many conventional materials change. This is usually because nanoparticles have a larger surface area per weight than larger particles, making them more reactive to other molecules. The study on nanoparticles are flourishing in the recent years with its numerous application in industries such as biomedical, optical and engineering fields, prompting researchers to develop ways to include nanoparticles for industrial application. However, it is quite difficult to obtain a stable dispersion of nanoparticles in the base fluid, which makes it important to understand the interaction that occurs between particle-particle and particle-liquid in order to prepare a stable fluid. Due to the attractive and repulsive forces on the particle surface, it generates different kinds of interactions among the particles involved. The most dominant forces would be the attractive molecular
forces, which causes particles to clump together, which is termed as agglomeration. Hence, these agglomerates reduces the stability of the nanofluids prepared.

Particles that were dispersed in a base fluid have a tendency to form larger aggregates and adhere together which may produce settlement due to gravitational forces which is also named as colloidal dispersion. A colloid can be liquid-liquid mixture or solid-liquid mixture. These would affect the stability of the prepared nanofluids. In order to determine the stability of nanofluids, one of the widely used methods by different researchers in literature is the sedimentation analysis or sediment ratio of nanoparticles [1–4]. This can be obtained by observing the rate of aggregation of the nanoparticles suspended in the base fluid. Sedimentation analysis is one of the method in which many researchers opt for in order to obtain the stability of the nanofluids that they prepared. It was proven to be one of the reliable method in determining a stability of a dispersion [5].

In general, the rate of aggregation is determined by the probability of cohesion and the frequency of collisions in fluids. A theory for colloidal stability were developed by Derjaguin, Landau, Verway and Overbeek (DLVO) [6–8]. The theory suggests that the stability of a particle in solution is determined by the sum of van der Waals attractive and electrical double layer repulsive forces that exist between particles as they approach each other due to the Brownian motion they are undergoing. To reduce these forces, the morphological characteristic of nanofluids are highly affected by the intensity of stirring and sonication time during its preparation.

Most of the researchers tend to add surfactant during preparation of nanofluids, which acts as dispersant, and reducing the agglomeration of nanoparticles in base fluid. In addition, the preparation method and choice of nanoparticles used also could highly affect the stability of nanofluids. Recently, many researchers utilized ultrasonic and stirring technique for preparing nanofluids in either one-step or two-step method [9–15]. Ultrasonic technique is commonly used to agitate and disperse nanoparticles into finer particles in the base fluid. S. Suresh et al. [16] studied the stability of spherical Al2O3 nanoparticles in deionized water after 6 hours of sonication at a concentration of 0.1 vol%. The author stated that the prepared nanofluids were stable for several weeks without any visible sedimentation. Xia et al. [17] reported that when using two different types of surfactant under the same ultrasonic intensity during preparation of Al2O3 nanofluids, the suspension with non-ionic surfactant shows better dispersion and longer stability as compared with anionic surfactant after 48 hours. Verma and Tiwari [18] in their studies revealed that by using ultrasonic method only, the prepared SiO2 nanofluids contain some agglomeration and lumping of nanoparticles. Despite that, the agglomerated lumps were in the nanosize range. In addition, Leena and Srinivasan [19] reported that the ultrasonic velocity increases with an increase in concentrations of TiO2 nanofluids. Hence, it shows that with an increasing concentrations of nanoparticles used, a much greater agitation is needed to overcome the agglomeration of particles in the base fluids.

Other than focusing on single type of nanoparticles, numerous study on hybrid nanofluids is becoming more common, with a vast mixture of different types of nanoparticles [20–22]. Maddah et al. [23] analyzed the stability of hybrid Al2O3-TiO2 nanoparticles in distilled water after 5 hours of sonication. The author found that the nanoparticles were homogenously mixed in the base fluid and it had sufficient stability. Moldoveanu et. al [24] studied on the thermal performance of stabilized Al2O3-SiO2 nanofluids prepared using one-step method and 1 hour of sonication. They reported that the prepared nanofluids were found to be relatively stable for about ten days. Although there are many reports available on the preparation method of nanofluids, none of them focused on finding the most suitable method to prepare stable hybrid nanofluids. In the present work, an attempt has been made to study the stability of hybrid Al2O3-SiO2 nanofluids in distilled water prepared using two-step method with different mechanical stabilizing technique.

2. Methodology

2.1 Preparation of nanofluids

The nanopowder used in this experiment were (1) Aeroxide Alu C (Aluminium Oxide, Al2O3) particles with \(d_p\) of 13nm and (2) Aerosil 90 (Silicon Dioxide, SiO2) particles with \(d_p\) of 20nm, obtained from
Aerosil Corporation. In the present experiment, the stated nanoparticles were mixed to achieve a hybrid Al$_2$O$_3$-SiO$_2$/water nanofluids of 50:50 vol% ratio which results in a concentration of 0.001, 0.01 and 0.1 vol%. The three concentration variations, $C = 0.001, 0.01$ and $0.1\text{ vol}\%$ were defined as low (L), medium (M) and high (H) concentration respectively.

Using two-step method, the nanoparticle powder were dispersed into 150ml distilled water inside a beaker. Then, 25ml of the nanofluids were transferred into 5 different test tubes. Successively, each test tube contains nanofluids with different preparation methods and labelled as (i), (ii), (iii), (iv) and (v) where test tube (i) represents the nanofluids that was sonicated for 1 hour, test tube (ii) is the nanofluids prepared using 1 hour of magnetic stirring while the nanofluids in test tube (v) did not undergo any stabilizing techniques. As for test tubes labelled (iii) and (iv), the nanofluids were prepared by implementing both magnetic stirring and sonication method. The nanofluids in test tubes (iii) and (iv) undergoes 0.25 and 0.5 hour of magnetic stirring using magnetic stirrer (FAVORIT Stirring Hotplate) respectively. The nanofluids were then sonicated for 1 hour using an ultrasonic agitator (Bransonic® CPXH Ultrasonic Bath). All of the nanofluids prepared were milky in colour with a darker gradient for higher concentrations. Table 1 presents the preparation method that the author used for this study.

| Nanoparticle | Concentration (vol%) | Method                        | Types   |
|--------------|----------------------|-------------------------------|---------|
| Al$_2$O$_3$-SiO$_2$ | 0.001                | 1 hour ultrasonic             | L(i)    |
|              |                      | 1 hour magnetic stirrer        | L(ii)   |
|              |                      | 0.25 hour magnetic stirrer + 1 hour ultrasonic | L(iii) |
|              |                      | 0.5 hour magnetic stirrer + 1 hour ultrasonic | L(iv)  |
|              |                      | None                          | L(v)    |
| Al$_2$O$_3$-SiO$_2$ | 0.01                 | 1 hour ultrasonic             | M(i)    |
|              |                      | 1 hour magnetic stirrer        | M(ii)   |
|              |                      | 0.25 hour magnetic stirrer + 1 hour ultrasonic | M(iii) |
|              |                      | 0.5 hour magnetic stirrer + 1 hour ultrasonic | M(iv)  |
|              |                      | None                          | M(v)    |
| Al$_2$O$_3$-SiO$_2$ | 0.1                  | 1 hour ultrasonic             | H(i)    |
|              |                      | 1 hour magnetic stirrer        | H(ii)   |
|              |                      | 0.25 hour magnetic stirrer + 1 hour ultrasonic | H(iii) |
|              |                      | 0.5 hour magnetic stirrer + 1 hour ultrasonic | H(iv)  |
|              |                      | None                          | H(v)    |

2.2 Sedimentation measurement
In this experiment, sonication time were set to 1 hour, while the stirring time were set to 0.25 and 0.5 hour. The Al$_2$O$_3$-SiO$_2$/water hybrid nanofluids were observed for 0, 12, 24 hour and 2 weeks after its preparation. The height of the sedimentation of the nanoparticles were observed by using photographic capturing of the prepared nanofluids in a glass test tube of 30ml volume. There were no manipulation of the test tubes during the observation process and the evaporation of the prepared nanofluids were prevented by using a screw-type cap test tubes (KIMAX).
3. Results and discussion

Based on figure 1, the test tubes were arranged starting from the left with test tubes labelled (i)-(v) consecutively. The configurations were the same for each concentrations. Figure 1(a) shows the sedimentation observations for the Al2O3-SiO2/water hybrid nanofluids at 0.001 vol%. After 2 weeks of observations, only slight agglomeration could be noticed in all of the test tube. Due to the small amount of nanoparticles used for this concentration, there is nearly no concluded observation could be drawn since the transparency of the nanofluids is similar to the distilled water used.

At concentration of 0.01 vol%, it can be seen that the stability of the nanofluids in test tube M(i) deteriorates after 6 hours with less nanoparticles suspended in the fluid, and hence producing a clear layer as shown in figure 1(b). From the observation, it can be seen that the sample in test tube labelled M(iii) and M(iv) had the highest stability with less settlement of nanoparticles at the bottom of the test tube. After 12 hour of preparation, clear layer started to form on all methods of preparation, but with the least formation of clear layer can be seen in test tube labelled M(iii) while the most visible layer could be seen in test tube M(i) as shown in figure 1(b). At 2 weeks of preparation, test tube labelled M(iii) had the highest colloidal stability, with lowest amount of sedimentation and clear layer formed. For concentration 0.01 vol%, it can be concluded that by implementing sonication and stirring method, it significantly improves the stability of the nanofluids prepared.

Figure 1(c) shows the sedimentation analysis for 0.1 vol% of the prepared hybrid nanofluids. After 12 hour of preparation, all of the nanofluids showed signs of slight phase separation, with the formation of clear layer in the test tube. Even though test tube H(ii) showed little to no signs of clear layer, it has the most amount of sedimentation, and becomes unstable. At 24 hours after preparation, it can be seen that the phase separation occurs more vigorously during this period. The clear layer of fluids appear to be more visible in each method of preparations, with test tube H(i) has the most obvious clear layer, followed by H(v), H(iv), H(iii) and H(ii). After 2 weeks, pictures are taken and it is found that settling velocity of nanoparticles prepared using both stirring and sonication is much lower than the others, as seen in test tube labelled H(iii) and H(iv). Clear layer is still visible for the samples, but the ratio of sedimentation is much lower than that of sample prepared without using both methods. In conclusion, the highest stability of dispersion of nanoparticles can be achieved by using the stirring and sonication method.

4. Conclusions

In this study, the stability of Al2O3-SiO2 hybrid nanofluid of 50:50 ratio were observed by using sedimentation analysis. The nanofluid were prepared using two-step method to achieve a concentration of 0.001, 0.01 and 0.1 vol%.

- It can be concluded that the dispersion stability of hybrid nanofluids prepared by applying magnetic stirrer prior to sonication had demonstrated the lowest height of sedimentation despite of the concentration of hybrid nanoparticles used. Even after repetitive experimentation, the results obtained for sedimentation remains the same.
- Method (iii) which is using 15 minutes magnetic stirring and 1 hour of sonication is considered sufficient in producing a stable nanofluids. This can be proven by the smaller aggregates of particles, which reduces the settling rate of the nanoparticles to gravity and hence produces the optimal aggregation of nanoparticles.
Figure 1. The prepared hybrid Al₂O₃-SiO₂ nanofluids of 50:50 ratio at (a) 0.001 vol%, (b) 0.01 vol% and (c) 0.1 vol% using different preparation method.
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