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Investigation on stability of *tri*-hybrid nanofluids in water-ethylene glycol mixture

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Abstract. Nanofluid is a suspension of liquid containing metal or non-metallic nanoparticles of typical size (1-100 nm) dispersed into the base liquid. Hybrid or composite nanofluids is considered an extension of research work for single nanofluids, which can be carried out through a combination of two or more different nanoparticles - either in mixed or dispersed composites in liquids. The objective of this study is to investigate the stability of *tri*-hybrid nanofluids suspended in Water-Ethylene Glycol (EG) mixture. The *tri*-hybrid nanofluids were prepared at a volume concentration of 0.05 to 0.3% using the two-step method. Three types of nanoparticles used namely Al₂O₃, TiO₂ and SiO₂, and dispersed in a base fluid of water/EG. The investigation on the stability of the hybrid nanofluids in the present study is conducted through UV-Vis, zeta-potential, sedimentation and micrograph observation. The findings from the investigations on the visual stability of sedimentation show that the differences in concentration (0.05, 0.1, 0.2, 0.3%) have been low in day 14. It was found that 10 h sonication time is the most suitable period for sonication to obtain a stable suspension. Comparison of data concentration ratio to sedimentation for single, hybrid and *tri*-hybrid nanofluids presents *tri*-hybrid nanofluids remains stable with a concentration ratio of 80%. Zeta potential evaluation conducted for the *tri*-hybrid nanofluids obtained the value of 25.1 mV in the classification of good stability. It can be concluded that the *tri*-hybrid nanofluids were successfully prepared and achieved good stability.

1. Introduction

Nanofluid is a suspension of liquid containing metal or non-metallic nanoparticles of typical size (1-100 nm) dispersed into the base liquid. In 1995, the concept of nanofluids was first introduced by Choi et al. [1]. This new method was proven to increase the heat transfer by improving the thermo-physical properties of the nanofluids. Nanofluids are known for the application in the heating and cooling process. The main cooling process is an important part of industrial applications such as power plants, chemical processes, microelectronics, transportation, automotive cooling systems etc. [2, 3]. The existence of solid particles leads to interesting characteristics in the fundamental thermo-physical properties of nanofluids. Thermal conductivity, viscosity, density and stability have been investigated in recent years by many researchers [4-8]. The method of nanofluids preparation is important to minimize the agglomeration of the nanoparticles, hence improve the stability. The most common process used in nanofluids preparation are one-step and two-step methods. The one-step method is the process of synthesizing nanoparticles and simultaneously dispersing them in a base liquid. However, this method is not practical for industry, which only applies to low vapor pressure liquids. Another method of nanofluid preparation known as the two-step method. There are two processes in this method, namely



(i) the synthesis of nanoparticles in powder form (ii) spreading the nanoparticles into the base liquid to form a stable and homogeneous solution [6, 9]. Most nanofluids that used oxide particles and carbon nanotubes were produced through a two-step method [10]. The two-step method is preferable for production of nanofluids in a large scale and thus applicable for the industry. However, the challenge of using the two-step method is that agglomeration and nanoparticles tend to settle quickly [11]. The two-step method is the most dominant method compared to the one-step method for nanofluid preparation.

Several recent studies discuss the topic of hybrid or composite nanofluids [12, 13]. Hybrid or composite nanofluids is considered an extension of research work for single nanofluids, which can be carried out through a combination of two or more different nanoparticles - either in mixed or dispersed composites in liquids [14]. Composite or hybrid materials are elements that combine chemical and physical properties. The aim of synthesizing hybrids or nanofluid composites is to improve the properties of single nanoparticles in which a better increase in thermal properties or rheological properties can be achieved. Hybrid nanofluid is expected to achieve good thermal performance when compared to a single of nanofluid [15].

Nanofluid stability and nanoclusters size affect parameters in thermal conductivity [16, 17]. Nanofluid stability is defined as nanoparticles resistance to aggregation. Factor such as the Van der Waals attractions cause aggregation, which results in the formation of nanoclusters contained in nanofluids [18-21]. The formation of nanoclusters depends on the size, which causes the settlement of particles in the nanofluids. The stability of nanofluid is very important for its application; there is a little study on estimating the stability of the suspension. UV-Vis spectrophotometric measurements have been used to quantitatively characterize the colloidal stability of the dispersions [18]. It can be applied to all base fluid, while zeta potential analysis has a limitation of the viscosity of base fluid [19].

Therefore, the objective of this study is to investigate the stability of *tri*-hybrid nanofluid suspended in W/EG. This study will help researchers to get an idea about the effect of nanoparticle stability on hybrid nanofluids, which will encourage researchers to apply nanoparticles in various types for cooling systems.

2. Methodology

2.1. Material

The preparation of *tri*-hybrid nanofluids involved three different types of single nanofluids namely Al_2O_3 , TiO_2 and SiO_2 mixed together and dispersed in the base fluid of water/EG mixture. All the single nanofluids were procured from US Research Nanomaterials, Inc. The respective nanoparticles size for Al_2O_3 , TiO_2 and SiO_2 are 13, 50 and 30 nm with a purity of 99.8%, 99% and 99.99%. The properties of each nanoparticle are given in Table 1. The base fluid used in the present study was a mixture of water and EG at a ratio of 60:40 (vol.%). The properties of Ethylene Glycol is presented in Table 2. The nanoparticle size characterization of the *tri*-hybrid nanofluid by field scanning electron microscope (FESEM) technique. The FESEM image for nanoparticles shown in Figure 1.

Table 1. Properties of Al_2O_3 , TiO_2 and SiO_2 nanoparticles [22, 23]

Properties	Al_2O_3	TiO_2	SiO_2
Molecular mass, g mol^{-1}	101.96	79.86	60.08
Average particle diameter, nm	13	50	30
Density, kg m^{-3}	4000	4230	2220
Thermal conductivity, $\text{W m}^{-1} \text{K}^{-1}$	40	8.4	1.4
Specific heat, $\text{J kg}^{-1} \text{K}^{-1}$	773	692	745

Table 2. Properties of Ethylene Glycol (EG) [24]

Properties	EG
Vapour pressure, mmHg at 20 °C	0.08

Boiling point, °C	195–198
Melting point, °C	-13
Density, g ml ⁻¹ at 25 °C	1.113

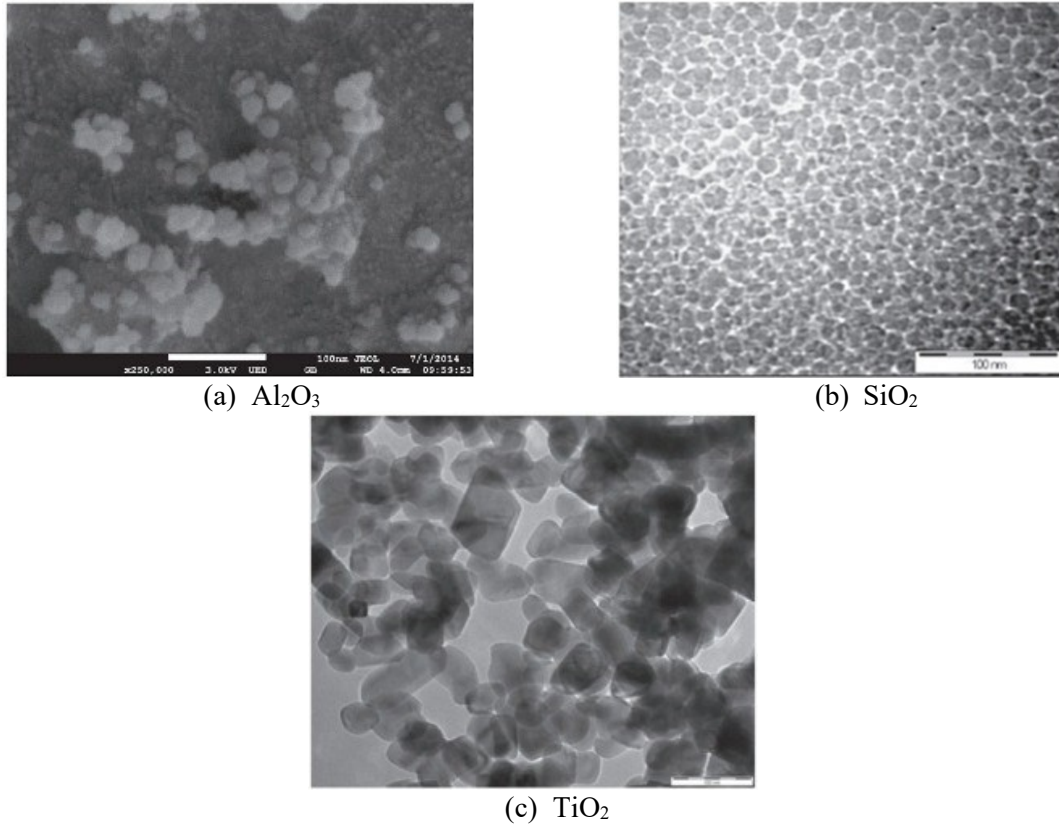


Figure 1. TEM Image for nanoparticles [22, 23, 25]

2.2. Preparation of tri-hybrid nanofluids

The two-step method is used for the preparation of *tri*-hybrid nanofluids. The *tri*-hybrid nanofluids were prepared by mixing all three single nanofluids (Al₂O₃, TiO₂ and SiO₂) together, undergo a mixing and sonication process. The preparation of the nanofluids was initially started with the calculation of the required volume according to the concentration. In the present study, the *tri*-hybrid nanofluids were prepared at a volume concentration of 0.05, 0.1, 0.2, and 0.3%. The nanofluids were first prepared at the highest concentration, 0.3% and then diluted to lower concentration.

The single nanofluids Al₂O₃, TiO₂ and SiO₂ were supplied in a water suspension with weight concentration of 20, 40, 25 % for Al₂O₃, TiO₂ and SiO₂, respectively. Eq. (1) [25] is used to convert from weight concentration to volume concentration. The dilution from higher volume concentration to lower volume concentration utilised the Eq. (2) [26] by adding the base fluid (ΔV).

$$\phi = \frac{\omega \rho_w}{\frac{\omega}{100} \rho_w + \left(1 - \frac{\omega}{100}\right) \rho_p} \quad (1)$$

$$\Delta V = (V_2 - V_1) = V_1 \left(\frac{\phi_1}{\phi_2} - 1 \right) \quad (2)$$

All single nanofluids were mixed together at a volume ratio of 1/3:1/3:1/3 to form a hybrid nanofluid. Total volumes of 100 mL were prepared for each concentration of the hybrid nanofluids. The combined solution from the three single Al₂O₃, TiO₂ and SiO₂ nanofluids were mixed together using magnetic stirrer for 120 minutes. Then, the solution was undergone sonication process using the ultrasonic bath to enhance the stability.

2.3. Stability of tri-hybrid nanofluids

The investigation on the stability of the *tri*-hybrid nanofluids in the present study is conducted through visual observation, measurement of UV-Vis Spectrophotometer and zeta potential. The sedimentation through visual observation was conducted up to 14 days. Nanofluid will be considered stable when the concentration is constant [27]. Previously, Azmi et al. [25] also used the same method to observe visual sedimentation of the prepared nanofluids. The UV-Vis was conducted for 10 days (250 h) by varying the sonication time. The wavelength of the UV-Vis spectrophotometer is set at 900 nm following the study by Hamid et al. [23]. The UV-Vis measures the absorption and light intensity of scattering nanofluid by comparing intensity level with the base fluid. The absorbance ratio of sonication times is different during sedimentation time at a constant wavelength (λ) of 900 nm. The stability evaluation by UV-Vis was also used by previous studies [29, 30]. The measurement by zeta potential was conducted using Zetasizer Nano ZS (Malvern Instruments Ltd., GB) [31].

3. Results and Discussion

3.1. Stability characterization of tri-hybrid nanofluids with UV-Vis Method

The observation of absorbance for volume concentration from 0.05 to 0.3% is shown in Figure 2. The absorbance of nanofluids is linearly increasing with the increase of volume concentration. This trend is in agreement with the Beer-Lambert Law, which is the value of the absorbance is equivalent to concentration [28].

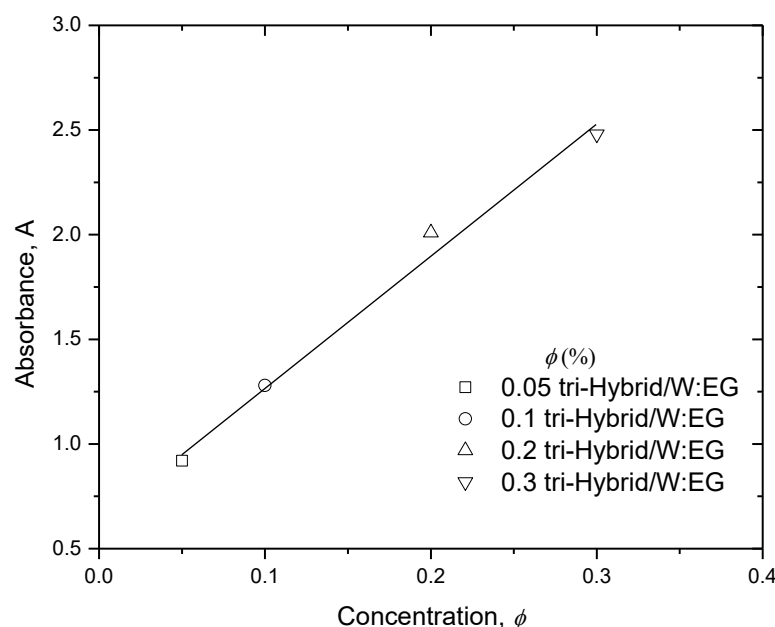


Figure 2. UV-Vis spectrophotometer linear relation graph between absorbance and *tri*-hybrid nanofluids concentration

Figure 3 demonstrates the concentration ratio of 0.1% volume concentration of the hybrid nanofluids for six different sonication hour. The ideal absorbance ratio is one (100%) which present the ideal stability of the fluid. The concentration of 0.5, 1 and 2 sonication hour start to decrease after 24 h and keep decreasing until 10 days (240 h). While the 5 h and 10 h sonication time remain at good

concentration ratio value at about (i.e. 70~80%) until 10 days later. From this figure, it can be seen that the 10 hour sonication time shows the best absorbance ratio compared to others. Thus, the preparation of the hybrid nanofluids in the present study used 10 h for the sonication process.

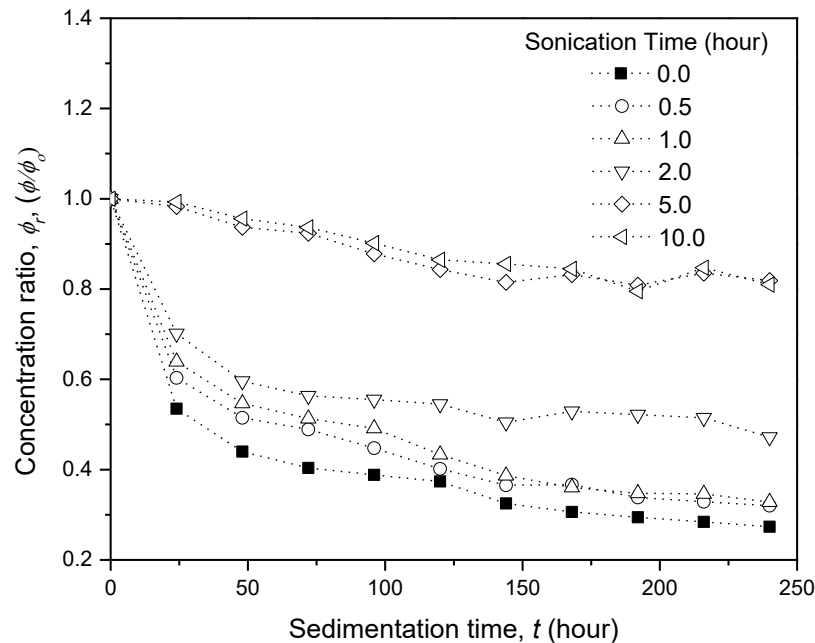


Figure 3. The concentration ratio of *tri*-hybrid nanofluids for different sonication time as a function of time

3.2. Comparison with literature for the concentration ratio

The concentration ratio of tri hybrid nanofluids in the present study is compared with Hamid et al [23] and shown in Figure 4. The investigation by Hamid et al [23] used $\text{TiO}_2\text{-SiO}_2$ dispersed in water/EG mixture. The concentration ratio maintained at more than 70% after a sedimentation time of 240 hours compared to other sonication times. Hwang et al. [19] used multi-walled carbon nanotubes (MWCNT) in the base liquid in water. As shown in the graph, MWCNT nanofluids have poor stability. In this study, $\text{Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$ used in the base liquid mixture of EG/Water and use sonication time 10 hour. The findings of concentration ratio obtained 80% from 150 to 240 minutes. This condition is stable.

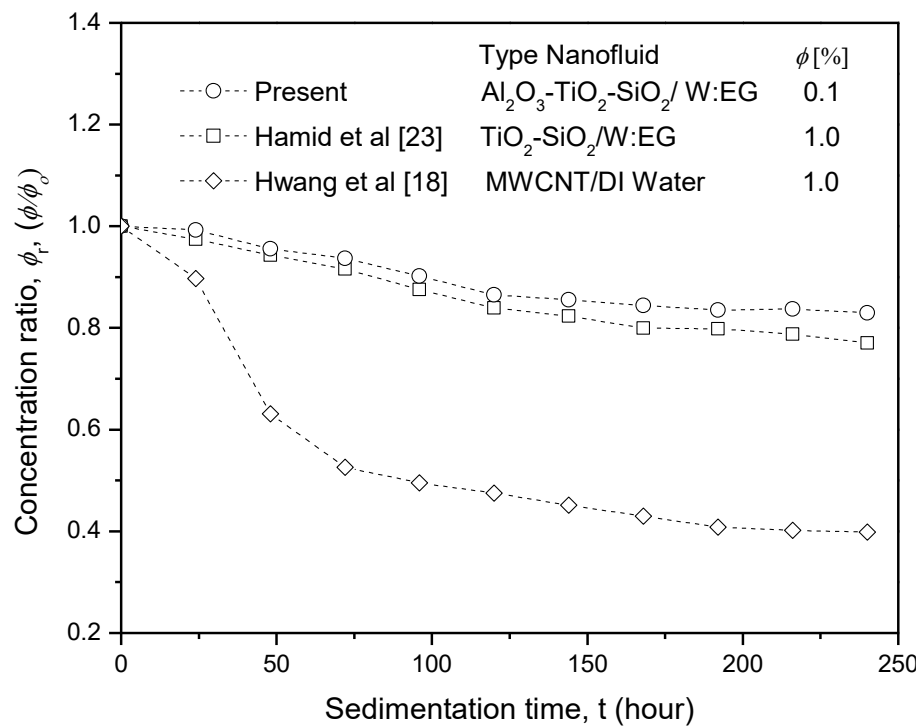


Figure 4. Comparison of concentration ratio with sedimentation ratio

3.3. Zeta Potential Evaluation

Zeta potential measurement is one of the most critical tests to validate the quality of nanofluids stability via a study of its electrophoretic behaviour [29]. Typically, accepted zeta-potential values summarized below in Table 3. Generally, a suspension with a measured zeta-potential above 30 mV (absolute value) is considered to have good stability [30]. This is one of the most common methods among the researchers to determine stability.

Table 3. Zeta potential and classification stability [30]

Zeta potential (mV)	Stability
0	Little or no stability
15	Some stability but settling lightly
30	Moderate stability
45	Good stability, possible settling
60	Very good stability, little settling likely

Zeta potential absolute value is used to show the agglomeration of $\text{Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$ nanoparticles in the EG/W. The higher the absolute value, the better the dispersion of the particles, hence better in term of stability. Zeta potential evaluation is a standard quantitative method for stability evaluation [31]. The zeta potential measurement for the *tri*-hybrid nanofluid recorded up to 25.1 mV. The result has then been compared with the classification of nanofluid stability based on the zeta potential of moderate stability value drawn up by Ghadimi et al. [30] as shown in Figure 5. The absolute value above 30 mV is desirable for good stability of the *tri*-Hybrid nanofluid. It evidently proved that the *tri*-Hybrid nanofluid is beyond the stable limit of 30 mV. Hence, the zeta potential evaluation has confirmed the stability of *tri*-hybrid nanofluid.

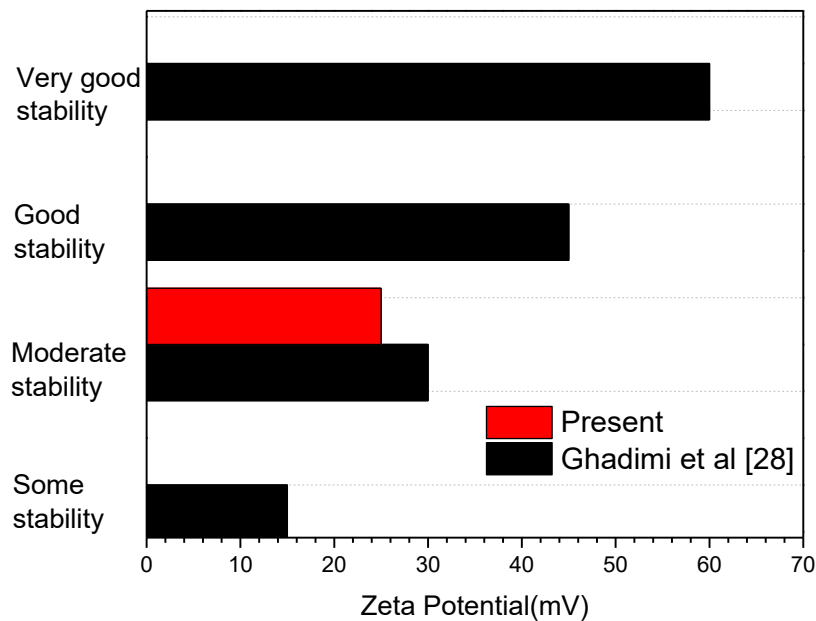


Figure 5. Zeta potential value of *tri*-hybrid nanofluid

3.4. Visualization effect

Figure 6 shows images of *tri*-hybrid nanofluid ($\text{Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$)-W/EG for a volume concentration of 0.05, 0.1, 0.2, 0.3%. *Tri*-hybrid nanofluid ($\text{Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$)-W/EG images were taken only after preparation and after 14 days. From Figure 6a, no sedimentation of particles observed after the nanofluids is prepared. The sedimentation of particles started to noticeable by day 7 afterward. After 14 days, the sedimentation can be clearly seen as shown in 6d. The sedimentation from this observation is affected by the gravity of the particle's falling motion in the tube.

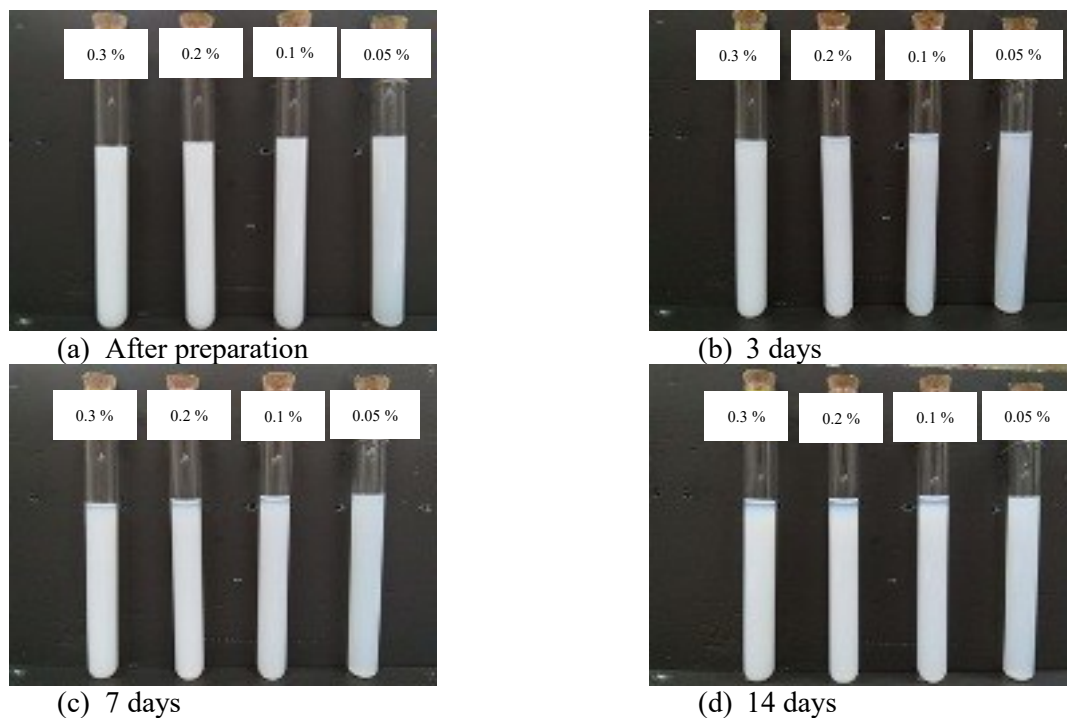


Figure 6. Sedimentation observation of *tri*-hybrid nanofluids: (a) after preparation, (b) 3 days, (c) 7 days, and (d) 14 days

4. Conclusions

Preparation of tri-hybrid nanofluids can be successfully prepared using a two-step method. Stability analysis of tri-hybrid nanofluids made by UV-Vis method that is stable up to 10 days after preparation with a 10-hour sonication time. The comparison of data concentration ratio to sedimentation for single, hybrid and *tri*-hybrid nanofluids presents *tri*-hybrid nanofluids remains stable with a value of 80%. Zeta potential evaluation conducted for the *tri*-hybrid nanofluids obtained the value of 25.1 mV in the classification of good stability. The sedimentation from this visual observation is affected by the gravity of the particle's falling motion in the tube on after 14 days. All methods prove that hybrid nanofluid containing three different metal-oxides (Al_2O_3 - TiO_2 - SiO_2) are combined together to form a stable solution.

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