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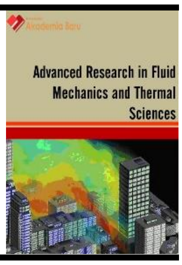

Experimental Investigation on the Stability of 40% Ethylene Glycol Based TiO_2-Al_2O_3 Hybrid Nanofluids

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ABSTRACT

This paper is presented to investigate experimentally on the stability of 40% ethylene glycol-based TiO_2-Al_2O_3 hybrid nanofluids. Recently, the research is more highlighted on the thermophysical-properties of nanofluids. Hence, the stability of the hybrid nanofluids thoroughly assessed in this research work. The study uses the two-step method for preparing 40% ethylene glycol-based TiO_2-Al_2O_3 hybrid nanofluids. The experiment is carried out for the various combination of mixture ratios including 20:80, 40:60, 50:50, 60:40 and 80:20 of TiO_2-Al_2O_3 nanoparticles with a volume concentration of 0.1%. The stability assessment of hybrid nanofluids is accomplished through visualisation effect, transmission electronic microscopic observation, UV-Vis spectrophotometry and zeta potential value from particle size analyser. The findings show the optimum mixing ratios of TiO_2-Al_2O_3 nanoparticles in terms of stability which is further confirmed by Zeta potential and absorbency from UV-Vis spectrophotometry. The results from the study reveal that 80:20 ratio of TiO_2-Al_2O_3 nanoparticles possesses the best uniformity for over 21 days without any noticeable settlement of particles in the colloidal suspension. Moreover, 50:50 and 60:40 ratios show modest stability for almost two weeks while 20:80 and 40:60 ratios of TiO_2-Al_2O_3 nanoparticles show minimum stability along with rapid sedimentation in the dispersion. However, it is also evident that the optimum mixing ratio of TiO_2-Al_2O_3 hybrid nanofluids has excellent stability which will lead to further study on the thermal properties of nanofluids and finally motivate engineers to the real-life application of nanofluids.

Keywords:
Hybrid nanofluids; stability; sedimentation photograph; ultravoilet-visible spectral analysis; zeta potential

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1. Introduction

The advancement with the thermal engineering, the emergence of improved heat transfer fluids is of keen interest of recent researches to enhance heat transfer in various industries such as manufacturing, biomedical, microelectronics, aerospace and others. The most important parameter which affects the heat transfer efficiency of fluids is the thermal conductivity of fluids. Thus, the low thermal conductivity of traditional fluids such as water, ethylene glycol, oils and others is the initial shortcoming in the efficient heat transfer process. However, it is evident from numerous researches that the inclusion of nano-sized particles into the base fluid has superior thermal properties than traditional fluids [1-3]. Thus, the development of nanofluids, which are composite materials or colloidal dispersion of nano-sized solid particles, tubes, rods or fibres into the base fluids [4-7], has attracted the attention of the recent scientists. The addition of solid particles also boosts up the load-carrying and anti-wear capacity of base fluids besides showing minimum friction [8-12]. Moreover, the heat transfer system can simplify through a size reduction of the system because of the application of ultrafine solid particles [13]. Despite having all these superior properties, the stability of nanofluids is still a challenge in the practical field of nanofluids because the nanoparticles tend to be agglomerated or sedimented causing pressure drops because of robust Van der Waals attraction among particles [14,15]. It leads to the reduction of stability as well as thermal properties of nanofluids [16-18] as stability directly correlated with the thermal properties [19,20]. Hence, the stability of the nanofluids is an essential factor for sustaining the enhanced thermal features [19,21]. So, stability analysis is the prior most work in the field of nanofluids.

To achieve homogeneity or stability of nanofluids, the synthesis method of nanofluids can play an important role. There are two standard synthesis processes of nanofluids- one-step and two-step methods. The process of synthesization and suspension of nanoparticles into the base fluids occurs concurrently in the one-step process [22,23]. On the other hand, in the two-step method, there are two processes for the preparation of nanoparticles using various synthesis techniques and then dispersion of particles into the fluid [22,24]. Numerous researchers used a one-step method for preparation of multiple nanofluids such as Fe_3O_4-CNT/ distilled water [25], CuO/ distilled water [26], Cu/ diethylene glycol [27]. The one-step method is employed for achieving uniform dispersion of nanoparticles and for sustaining enhanced thermal properties [28], but this method is not preferred at the industries due to its higher production cost and limited application of this process for only fluids having low vapour pressure [29]. On the contrary, the two-step method is utilised mainly in various industries for the synthesis of nanofluids [15]. Additionally, most of the researchers use this method during the preparation of nanofluids [30-32]. Choi and Eastman [4] especially suggested about two-step synthesis technique for oxide base nanoparticles rather than nanofluids based on metal particles. However, the agglomeration of nanoparticles is a critical challenge during employing this method [28]. Stability of nanofluids observed as the ability to resist sedimentation or agglomeration. Strong Van der Waals attraction force is responsible for the nanoclusters formation in the nanofluids [33-36]. Achieving stable nanofluids is a crucial need in the practical field of nanofluids. So, in this research, the stability analysis of 40% ethylene glycol (EG) based TiO_2-Al_2O_3 hybrid nanofluids is conducted for various ratios of nanoparticles.

Researchers used various methods for achieving stability of nanofluids such as, ultrasonication method [30,37], surfactant addition [3,38,39], pH modification [40,41], surface modification technique [42,43] etc. For instance, Elias, Mahbubul [37] found stability up to three days while preparing Al_2O_3 nanofluid using the two-step method. In case of water-EG based TiO_2-SiO_2 nanofluid, Hamid, Azmi [32] achieved 14 days stability period for 2 hours ultrasonication. Moreover, surfactant addition can also play an influential role in achieving stable and durable nanofluid [44,45]. Ouikhalfan, Labihi [3] used CTAB and SDS treated TiO_2 particles for preparing stable nanofluid and finally found stability for over two weeks with 15% and 10% enhancement of thermal conductivity for volume concentration of 1.25%. Distilled water-based Al_2O_3 nanofluid acted as a stable nanofluid after appropriate sonication with SDS surfactant addition reaching the zeta potential value almost ±30mV [40]. The use of surfactant should be avoided for high-temperature applications [46,47] because of properties degradation of surfactant at temperatures more than 60^0 C [48].

However, for achieving stability, this study follows the simplest ultrasonication method, and for the stability assessment, it uses transmission electronic microscopic (TEM) photograph showing the arrangement of the particles, UV-vis spectral analysis and zeta potential measurement for quantitatively characterise the uniformity of nanofluids. As well, the visual observation correspondingly used by taking sedimentation photograph of dispersion. Hence, a combination of various methods reconfirms the results of the study. Some researchers use more than one way to show the stability of nanofluids. For example, Li, Zhu [49] and Hamid, Azmi [50] use UV-vis and visual observation for aqueous copper dispersion and water-EG based TiO_2-SiO_2 nanofluids. Asadi, Asadi [1] only uses zeta potential analysis to represent the stability period of nanofluids. Finally, the result of the present research compared with other literature. Thus, this research will contribute to achieving a clear view about the effect of solid particles and also stability of hybrid nanofluids which may finally direct to the practical application of nanofluids in different fields successfully.

2. Methodology

2.1 Preparation Method of Nanofluids

The most dominant two-step preparation method is employed to prepare 40% EG (60:40 mixture ratio of water and EG) based TiO_2-Al_2O_3 hybrid nanofluids. In this study, six different mixing ratios of 20:80, 40:60, 50:50, 60:40, 80:20 of TiO_2 and Al_2O_3 nanoparticles at 0.1% volume concentration are used for synthesization of hybrid nanofluids without using any surfactant. All the nanoparticles are purchased from US Research Nanomaterials, Inc. The properties of nanoparticles given by the manufacturer are presented in Table 1. Various metal oxides such as TiO_2, SiO_2, CuO, ZnO, Al_2O_3 have better dispersibility in working fluids [51]. For instance, improved stability of TiO_2/water-EG [52], SiO_2/water [53], Al_2O_3/water-EG (80:20) [54], TiO_2-SiO_2/ water-EG [55] are reported. Besides, A_2O_3 have greater thermal properties compared with the mentioned oxide solid particles [1]. So, it is expected that the composite of TiO_2-Al_2O_3 nanoparticles will possess a good uniformity along with stimulating thermal properties. Eq. (1) is used to measure the nanoparticles volume concentration (ϕ) from weight concentration (ω) and Eq. (2) is used to achieve the required concentration through the dilution process. However, a specific amount of solid particles are dispersed into 40% of EG following six different mentioned mixing ratios. Firstly, the magnetic stirrer is used for 45 minutes to mix the nanoparticles into the base fluid uniformly. Again, the dispersion is undergone to the sonication process to break down the agglomeration of particles and to achieve a homogenous solution with greater stability. Figure 1 presents the preparation of 40% EG based TiO_2-Al_2O_3 hybrid nanofluids.

$$\varphi = \frac{\omega \rho_{\mathrm{bf}}}{\left(1\text{-}\frac{\omega}{100}\right)\rho_p + \frac{\omega}{100}\rho_{bf}} \tag{1}$$

$$\Delta \mathrm{V} = (V_2\text{-}\mathrm{V}_1) = \mathrm{V}_1\,(\varphi_1/\varphi_2\ \text{-}1) \tag{2}$$

where, ρ_p, ρ_{bf} are the density of nanoparticles and base fluid respectively and V is the volume of colloidal suspension.

Table 1
Properties of TiO_2 and Al_2O_3 nanoparticles

Characteristics	TiO_2	Al_2O_3
Purity (%)	>99	99.8
Colour	White	White
Average particle diameter (nm)	30-50	13
Molecular Mass (g mol^{-1})	79.86	101.96
Density (Kg m^{-3})	4230	4000
Thermal conductivity (W m^{-1} K^{-1})	8.4	40
Specific heat (J Kg^{-1} K^{-1})	692	773

(a) (b)

Fig. 1. Preparation of 40% EG based TiO_2-Al_2O_3 hybrid nanofluids (a) Magnetic Stirring (b) Ultrasonication

2.2 Stability of Nanofluids

Stability or the repulsive force to the settlement of nanoparticles in suspension has dominant effects on the attributes of nanofluids. This study conducts visual observation for 21 days for stability analysis of TIO_2-Al_2O_3 hybrid nanofluids. Visual inspection is also followed in previous investigations [50,55,56]. Besides, absorbance value is found through Uv-vis spectral analysis for specifying the stability condition of nanofluids. The UV-vis spectrophotometer analysis method is also used by other researchers [57-59]. Moreover, particle size analyser (Malvern Zetasizer Ultra- DKSH) is employed to measure the zeta potential of nanofluids. The transmission electron microscopic images of TiO_2-Al_2O_3 presents in Figure 2.

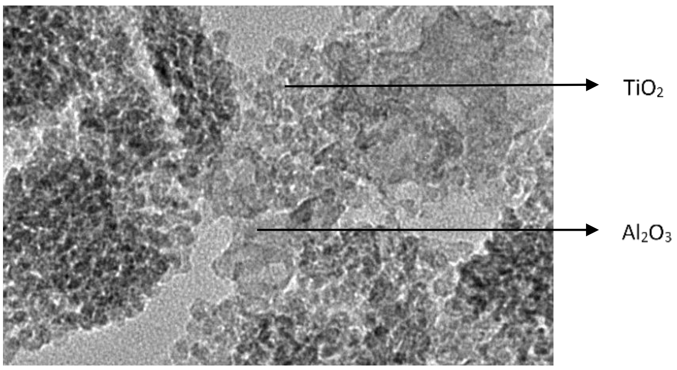

Fig. 2. TEM images of 40% EG based TiO_2-Al_2O_3 nanofluids at 0.1% vol. concentration

3. Results and Discussion

3.1 Sedimentation Photograph

The nanofluids are observed for one month, and images of the state of suspension are taken from time to time. From Figure 3(a), no settlement of nanoparticles is found after the nanofluids are synthesised. After two days, 20:80, 40:60, 60:40 ratios of TiO_2- Al_2O_3 containing samples start to settle down presented in Figure 3(b). After seven days, a few settlements of nanoparticles is also observed for the ratio of 50:50 (Figure 3(c)). Moreover, 80:20 ratio of TiO_2- Al_2O_3 nanofluid is found as stable for two weeks without sedimentation (Figure 3(c)). Finally Figure 3(d) represents the status of suspension after 21 days where 20:80, 40:60 ratios show a large amount of deposition followed by 60:40, 50:50 ratios of nanoparticles and 80:20 ratio shows very few amounts of deposition which may be due to gravitational force or the falling motion of nanoparticles [60].

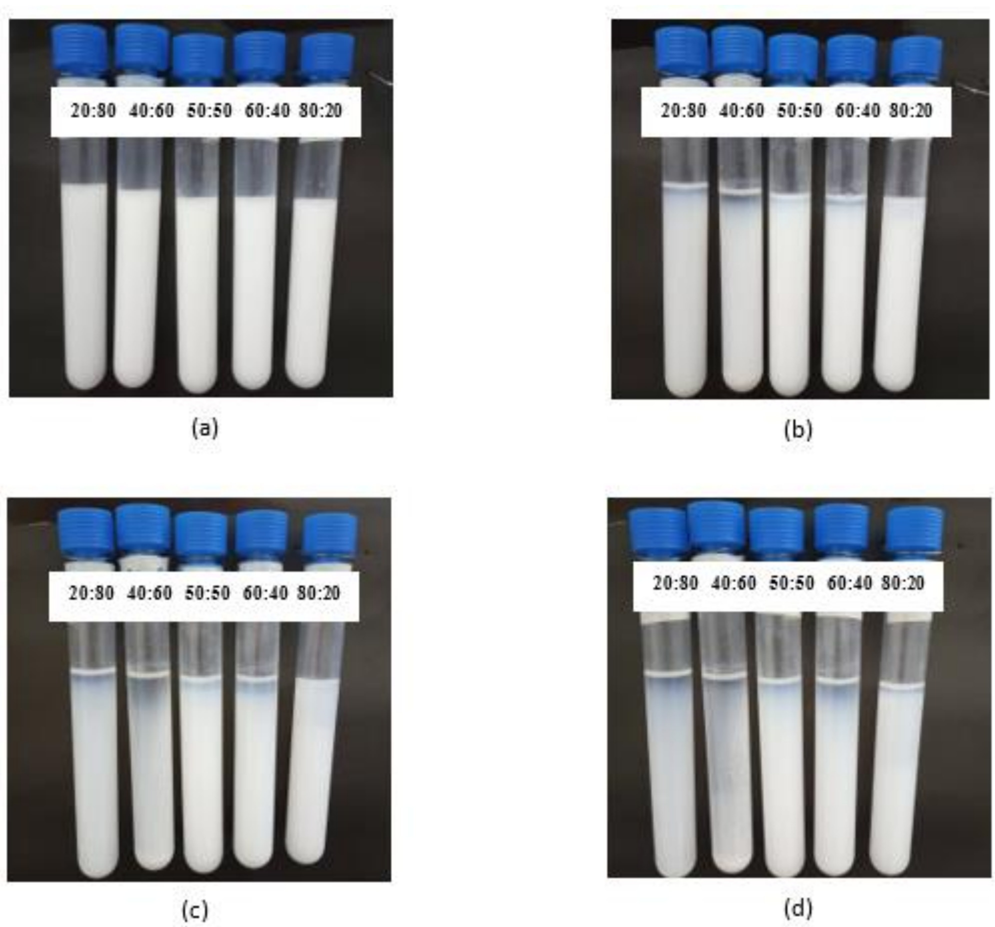

Fig. 3. Sedimentation photograph of TiO_2-Al_2O_3 hybrid nanofluids (a) Just after preparation (b) After 3 days (c) After 14 days and (d) After 21 days

3.2 UV-vis Spectral Analysis Method

The absorbance of the nanofluid is investigated in UV-vis spectrophotometer by comparing the light intensity between base fluid and nanofluid [48]. Yu and Xie [28] suggested the wavelength range from 190 to 1100nm for achieving the most authentic value. In this study, a constant wavelength of 903 nm selected through scanning the nanofluid in UV-vis spectrophotometer because the peak value of absorbance derived from this wavelength. The absorbance values of TiO_2-Al_2O_3 for mixing ratios of 20:80, 40:60, 50:50, 60:40 and 80:20 at 0.1% volume concentration are presented in Figure 4. The absorbance value is increasing proportionately with increasing TiO_2 particles and lowering Al_2O_3 nanoparticles. The trend of absorbance value aligned with Beer-Lambart Law [60,61]. Because absorbance value is linearly increasing responding with various mixing ratios of TiO_2-Al_2O_3. From Figure 4, it is discovered that the more the ratio of TiO_2, the more the value of absorbance, which leads to more stability. On the other hand, the more the ratio of Al_2O_3, the less the value of absorbance of nanofluid. The reason for this behaviour may be due to the strong van der Waals force among Al_2O_3 nanoparticles.

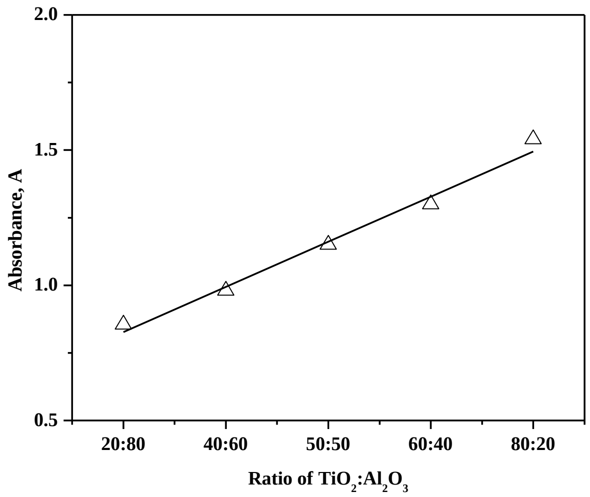

Fig. 4. The absorbance value of 40% EG based TiO_2-Al_2O_3 hybrid nanofluids for various mixing ratios at 0.1% vol. concentration

3.2 Zeta Potential

Zeta potential test is a critical method to assess the stability of nanofluid quantitatively. The electrostatic repulsive force between the suspension medium and static layer of fluid adhered to the particles [40]. The absolute value of zeta potential depicts the uniformity of colloidal suspension [28]. In general, the lower value of zeta potential expresses quick settlement of particles in the suspension [48]. However, zeta potential value with near to 30mV can be regarded as moderately stable [62]. Table 2 represents recognised zeta potential value with the stability state of colloidal suspension. However, this method has some limitation on the viscosity and concentration of fluid [15,33]. In this study zeta potential value is employed to present the sedimentation of TiO_2-Al_2O_3 nanoparticles in 40% EG. The higher the value, the better the nanofluid in terms of the uniformity of suspension. Figure 5 shows the absolute value of zeta potential for different mixing ratios of TiO_2-Al_2O_3 hybrid nanofluids. The maximum of the value of zeta potential is recorded up to 34mV for 80:20 ratio of TiO_2-Al_2O_3 while 50:50 and 60:40 ratios show the value of zeta potential near to 30mV (25mV and 23mV respectively). On the other hand, 20:80 and 40:60 ratios show little bit stability value of 17mV and 20mV, respectively. So it is evident that 80:20 ratio of TiO_2-Al_2O_3 nanofluid has good stability followed by 50:50 and 60:40 ratios which are moderately stable.

Although there are numerous researches in the field of nanofluids, the use of nanofluid is still in the research and development stage, not in practical application broadly. Various factors like agglomeration, settlement of nanoparticles, corrosion and wear potential need to be characterised as detail according to the shape and size distribution in the research to make the findings more reliable and useful.

Table 2
The state of stability of nanofluids against zeta potential value [62]

Stability state of Nanofluids	± Value of Zeta Potential (mV)
Excellent stability	60
Good stability	45
Moderate Stability	30
Lightly stable	15
No stability	0

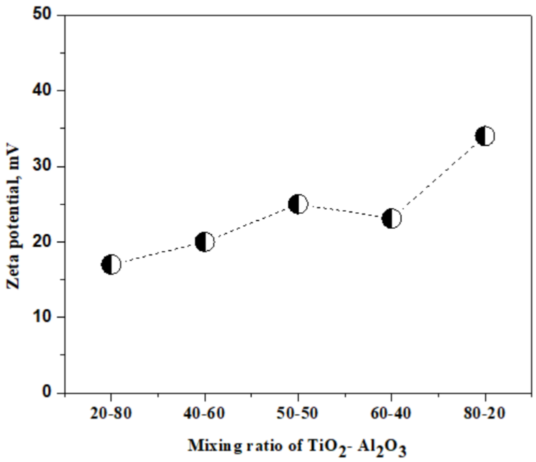

Fig.5. Zeta potential value 40% EG based TiO_2-Al_2O_3 hybrid nanofluids for various mixing ratios at 0.1% vol. concentration

However, the results from the previously published works of literature to date are adequate to get an idea about some magnitudes and also the trends in the enhancement of thermal properties of nanofluids [50,51], but still, some is lacking in the stability field of different types of nanofluids. This study will help researchers to achieve a clear idea about the stability of 40% EG based TiO_2-Al_2O_3 nanofluids for various mixture ratios. Hence, future research may be focused on the characterization of nanofluids and most importantly on the long-term stability of nanofluids which will be a significant contribution in the field of commercialisation of nanofluids.

4. Conclusions

In this research, the stability analysis of 40% EG based TiO_2-Al_2O_3 hybrid nanofluids shown after preparing using the most common two-step method. Ultrasonic vibration method employed in this study for stabilisation purpose of nanofluid without any surfactant and pH stabiliser. The time-dependent sedimentation observation method gives the qualitative idea of the stability of TiO_2-Al_2O_3 hybrid nanofluids. Moreover, the peak value of absorbance of the nanofluid, linear relationship of absorbance and various mixing ratio of TiO_2-Al_2O_3 hybrid nanofluids and also zeta potential value quantitatively confirms the stability analysis of studied nanofluids. This study reveals that the 80:20 ratio of TiO_2-Al_2O_3 hybrid nanofluids has the highest stability for over 21days without any apparent settlement of particles and followed by 50:50, 60:40, 40:60 and 20:80 ratio of nanofluids. Moreover, among all ratios 50:50, 60:40 are also moderately stable for two weeks with very few sedimentations whereas 20:80, 40:60 ratios show little stability with quick settle down of articles. Thus, 80:20, 50:50, 60:40 ratios are suitable to use in the practical field and stability period to enhanced with various treatment like surface modification, pH modification and if necessary additives addition. Future work will be carried out for sustaining the long term stability of different types of hybrid nanofluids which will contribute to practical and commercial applications.

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