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PREPARATION AND STABILITY ANALYSIS OF WATER BASED ALUMINIUM OXIDE NANOFLUIDS

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ABSTRACT
Nanofluid, as a brand new heat transfer fluid, is a colloidal suspension which enhances the heat transfer but its stability and durability are matters of concern for the industrial applications in the future. In this paper, Nanofluid based on water with aluminium oxide (γ-Al_2O_3) were synthesis by two step approach, previously prepared aluminium oxide nanoparticle dispersed by ultrasonication and analysis of stability is done to evaluate its appropriateness to be used in heat transfer application as a heat transfer fluid. Stability of nanofluids were analysed by varying the ultrasonication time under different particle concentration (0.015, 0.02, 0.05 and 0.1% by volume fraction) and by varying pH (1.5, 2.5, 4.15, 6.15, 10, 11.50, 12. 50). Zeta potential and sedimentation photo capturing were applied to analyze the sedimentation rate and stability of the prepared nanfluid. It was found that the stability of nanofluid take turn with pH and sonication time. The result shows that at an optimum pH and a optimum sonication time nanofluid shows excellent stability.

INTRODUCTION
Nanofluid is envisioned to describe a fluid in which nanometer-sized particles are suspended in conventional heat transfer basic fluids. Conventional heat transfer fluids, including oil, water, and ethylene glycol mixture are poor heat transfer fluids, since the thermal conductivity of these fluids play important role on the heat transfer coefficient between the heat transfer medium and the heat transfer surface. Therefore numerous methods have been taken to improve the thermal conductivity of these fluids by suspending nano/micro or larger-sized particle materials in liquids.

Preparation of nanofluids is key step to any experimental investigations. There are several method to prepare nanofluids, out of these the most important are single-step method and two step method. This single-step is the process of combining manufacturing of nanoparticle and synthesis of nanofluid in one step only as depicted in fig.2. In this process simultaneously making and dispersing of the particles in the basefluid avoid the drying, transportation, storage problems of nanoparticles.
Akoh et al. [1] has used one-step method in which nanoparticle of ferromagnetic metals (Fe, Co and Ni) were prepared by the vacuum evaporation onto a running oil substrate. Particle thus obtained were suspended in the oil. This method is involve direct condensation of Cu metallic vapor into nanoparticles by making contact with a flowing low vapor pressure liquid. Wagener et al.[2] did the modification of the above explained direct condensation method. Later on Eastman et al. [3] used it to developed a one-step physical method , in which vapor of Cu make contact with flowing low vapor pressure liquid ethylene glycol and condensed into the nano particle. Liu et al. [4] for the first time use chemical reduction method for synthesis of nanofluids containing nanoparticle of Cu in water. Cu nanoparticle is prepared in nitrogen atmosphere from copper acetate in water. Yu et al.[5] used phase transfer method for the development of kerosene based Fe_3O_4 nanofluids. Zhu et al. [6] developed a fast and efficient one step chemical method to prepare Cu nanofluids. This is a novel one-step method for preparing of copper nanofluids by the reactions of $CuSO_4.5H_2O$

with $NaH_2PO_2.H_2O$ in ethylene glycol under microwave irradiation. Lee et al. [7] prepared CuO nanofluids by laser ablation in liquids with the use of single pulse laser beam up to 8 hour. Lee et al.[8] prepared ethylene glycol based nanofluids containing TiO_2 nanoparticles by one-step pulsed wire evaporation method. The pulsed wire method is the kind of gas condensation method.

This two-step process is mostly used in the making of nanofluids, in this process the previously prepared nanopowder is get dispersed in the base fluid and then sonication is done as depicted in the fig.3. There is a lots of process to prepare the

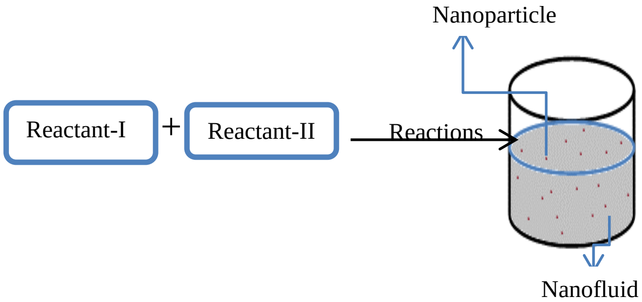

Figure 1. Schematic diagram of One-step process

nanopowder like inert gas condensation, chemical vapor deposition, sol-gel, vapor phase method etc. Dispersants and surfactant also added into the base fluid in small amount to increase the suspension stability.

Eastman[3] and Eastman et al.[9] used two-step method to produced alumina and copper oxide nanofluids. Lee et al.[10] and Wang et al.[11] also used the two-step method to produce alumina nanofluids. Masuda et al.[12] also did the same work i.e. prepared the nanofluids of aluminum by the two-step method. Murshad et al.[13] prepared their nanofuid by dispersing TiO_2 in water by two-step method. Carbon nanotube base nanofluid can be manufacture by this two-step method Choi et al.[14] reported the suspension of carbon nanotube by two-step method. Hu et al. [15] reported that aluminum nitride nanoparticles can be dispersed into ethanol with caster oil as dispersant so that suspension stability can be increased. The mixture then stirred by a high speed magnetic stirrer and then placed in ultrasonic homogenizer. Yu et al.[16] reported the dispersing of ZnO nanoparticles in ethylene glycol and then the mixture is stirred and ultrasonicated for better stability. Moosavi et al.[17] used the ethylene glycol and glycerol as the base fluid for ZnO nanoparticle and magnetic stirrer used to disperse the nanoparticle, Ammonium citrate used as a dispersant to obtain stable suspension and increasing the dispersing rate. He et al. [18] did the mixing of dry TiO_2 nanoparticle into the base fluid distilled water which have ph value of 11 and then the mixture is ultrasonicate.

Apart from the above explained method there are some methods also developed, some important from there are mention here. Feng et al.[19] developed an aqueous organic phase transfer method to prepare gold, silver, and platinum nanofluids which is an elemental but have effective nature. Yu et al.[20] used phase transfer method to prepare nanofluids having Fe_3O_4 nanoparticle and base kerosene, to make Fe_3O_4 particles compatible with kerosene oleic acid was successfully grafted onto the surface of particles.

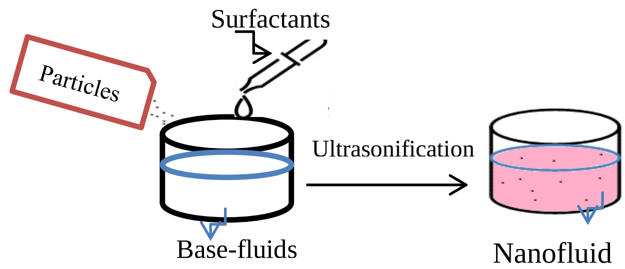

Figure 2. Two-step process of making nano-fluids

Though one step method avert the drying, transportation, storage problems of nanoparticles but it is not much used to make nanofluids. Generally two step method used to make nanofluids, it is simpler than one step method because nanoparticle either self-made or purchased. The most important advantages of two step method is that facile and speedy manufacturing of large volume nanofluids. However the challenge is to disperse nanoparticle fully and uniformly into base fluid, due to large and active surface area nanoparticles tend to agglomerate. Therefore, in order to prepare a stable nanosuspension sonication as well as addition of stabilizer or pH adjustment may be required.

In this work, preparation of water based aluminium oxide nanofluid has been done with different volume concentration of γ-Al_2O_3 in water. Stability was measured by settleing time and zeta potential with variation in sonication time and pH.

EXPERIMENT

Materials and apparatus

Aluminium oxide (γ-Al_2O_3) nanoparticles of 20 nm diameter were purchased from a US based company Nanoshel, properties of nanoparticle are given in table 1. All the chemicals utilized in this study were used as received without further purification. The beaker used in this process were cleaned by an ultrasonic cleaner in an ultrasonic bath. γ- Al_2O_3 nanoparticle were mixed with distilled water to prepare nanofluid of various volume concentrations by two step method. Mixture was shacked and stirred by hand for 10 minutes and then sonicate in probe sonicator (BMS-750T) purchased from Biomatrix company.

Afterwards, the pH of the nanofluid was change by using HCl and NaOH solution and then the nanofluids further processed for different process.

Table 1. Properties of Nanoparticle

Product Name/ Charecteristics	Alumina Powder Nano Grade (Gamma) (γ- Al_2O_3)
Color	White
Crystal form	Alpha
PH Value	6.6
Particle size	20 nm (gamma)
Al_2O_3 content	99.99%

X-ray differaction patterns (XRD) for γ-Al_2O_3 nanoparticle measured by using Bruker D8 X-ray diffractometer. Radial scan intensity versus scattering angle (2θ) recorded at room temperature with anode material copper and filament current 30 mA. To calculate the d-value Bragg's law was applied.

Stability of nanofluids determined by measuring zeta potential in Zetasizer Nano ZS90 purchased from Malvern Instruments Ltd., UK. Laser Doppler Micro-electrophoresis is the technique used to measure zeta potential. An electric field is applied to a solution of molecules or a dispersion of particles, which will then move with a velocity related to their zeta potential. This velocity is measured using a patented laser interferometric technique called M3-PALS (Phase analysis Light Scattering). This enables the calculation of electrophoretic mobility, and from this, the zeta potential.

RESULT AND DISCUSSION

X-Ray Diffraction (XRD)

The particles were characterized by XRD, the peaks shown in figure 3 indicate good crystallinity of aluminium oxide nanoparticle. The X-ray diffraction studies confirmed that the material is aluiminum oxide. The average crystal size of the nanoparticles is 20 nm which was calculated by Bragg's law shown in equation 1.

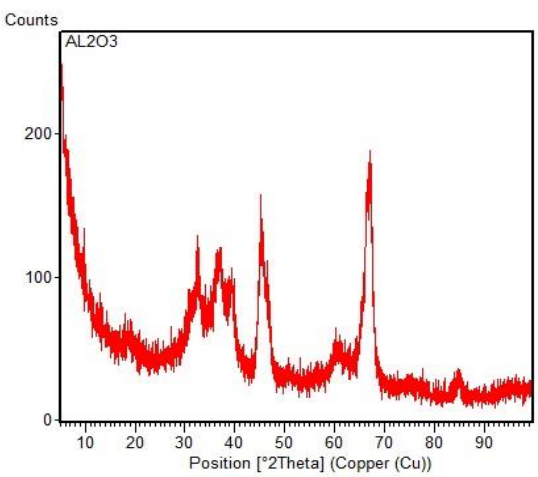

Figure 3. γ-Al_2O_3 pattern measured by XRD.

$$n\lambda = 2d \sin\theta \quad (1)$$

Where n is an integer, λ is the wavelength of incident wave, d is the spacing between the planes in the atomic lattice, and θ is the angle between the incident ray and the scattering planes.

Effect of sonication time on stability of aluminium oxide nanofluids

It was found that there is a optimum ultrasonication time for a particular concentration of nanoparticle in base fluid at which they shown higher stablity. For γ- Al_2O_3 nanofluid, the stability first increases wih ultrasonication time then after a optimum time it start decreasing as shown in the figure 4 and figure 5.

In ultrasonication, ultrasonic waves of 19 khz frequency passes from the nanofluid which causes vibration to the nanoparticles, this lead to the development of cavitation bubbles and growing during several cycles until they attain a critical state, which induces their implosion. This collapsion causes a very high local pressure and a very high temperature i.e locally extreme conditions, called hot-spot. These hot-spot responsible for the splitting up of particle agglomerations. The shock waves from the implosive collapsion and micro-streaming generated by cavitation oscillations combinatorially lead to dispersion effects [21]. That's why nanofluids more stable after proper sonication. But if the time of sonication exceed from the optimum time, it accelerate the nanoparticle with the increase of solution temperature leads to increment in the Brownian motion, which caused the collision of the nanoparticles, so agglomeration of nanoparticles occurred, resulting in decrement in stability.

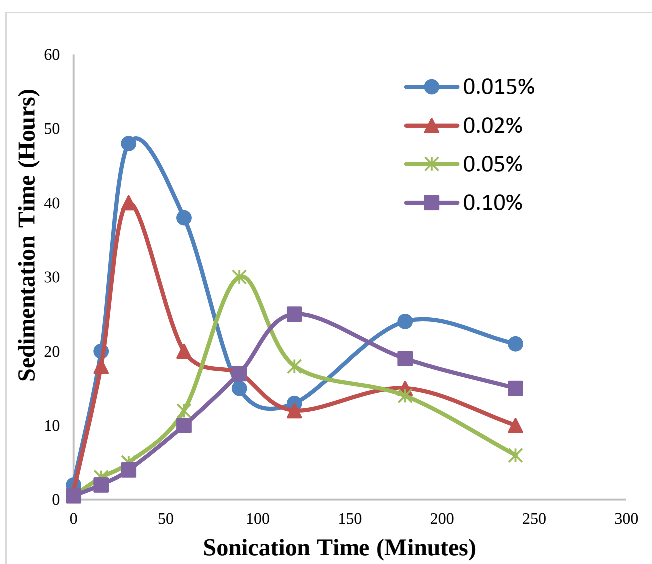

Figure 4. Sedimantation time for different volume fraction of γ- Al_2O_3-water nanofluid at different sonication time.

Figure 5 (A) & (B) Shows comparison between nanofluid sedimentation containing 0.015% (by vol.) nanoparticle at diffierent sonication time(30 min, 2 hr, 3 hr, 5 hr from right). (A) just after sonication. (B) after 25 hr of sonication.

Figure 5 (C) & (D) Shows comparison between nanofluid sedimentation containing 0.02% (by vol.) nanoparticle at diffierent sonication time(30 min, 1 hr, 2 hr, 5 hr from right). (C) just after sonication. (D) after 25 hr of sonication.

Figure 5 (E) & (F) Shows comparison between nanofluid sedimentation containing 0.05% (by vol.) nanoparticle at diffierent sonication time(30 min, 1 hr, 1hr 30 min, 5 hr from right). (E) just after sonication. (F) after 25 hr of sonication.

Figure 5. Photos of sedimentation after few hours of sonication

Effect of pH on stability by measuring Zeta potential

Generally a suspension with a measured zeta potential between -25 mV to 25 mV is highly unstable and above ±30 mV (absolute value) is considered to have good stability. Table 2 have the accepted values of zeta potential.

Table 2. Zeta potential values and associated suspension stability

Z potential (absolute value [mV])	Stability
0	No stability
15	Some but settling lightly
30	Moderate stability
45	Good stability
60	Very good stability

In this experiment the value of pH changed by adding NaOH and HCl solution into the nanofluid. The zeta potential varied as shown in the figure 6. For γ- Al_2O_3 nanoparticles value of the zeta potential is zero at 8.89 pH, which is called as isoelectric point, there the force of electrostatic repulsion between particles is not sufficient to overcome the force of attaraction between the nanoparticles and hence the nanofluid is least stable. By increment or decrement in pH leads to acquire more charge, so that electrostatic repulsion force between nanoparticles becomes sufficient to overcome the attraction and aggreagation between particles. As the electrostatic force increase, there is increment in particle-particle distance upto a level that it exceed the hydrogen bonding range, results in reduction of particles coagulation and settling, which increase the stability [22].

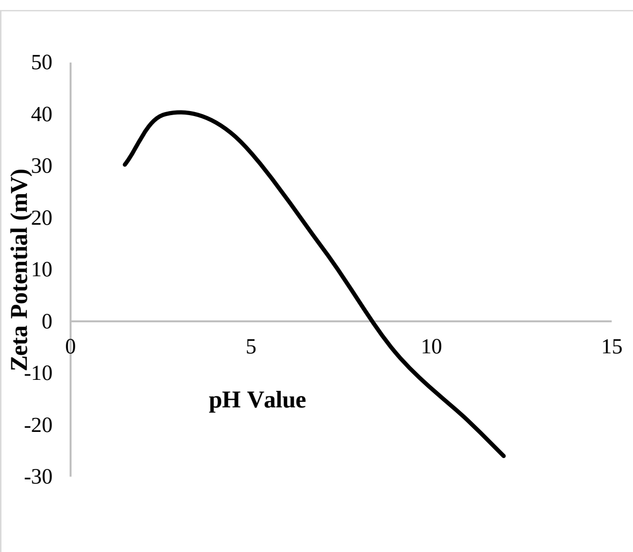

Figure 6. The development of Zeta potential of γ-Al_2O_3-water nanofluid with pH.

CONCLUSIONS

In this study, analysis of stability of water based γ- Al_2O_3 nanofluid has been conducted. Following conclusion can be drawn from the present study:

1. For different volume fraction of γ- Al_2O_3 nanoparticles in base fluid, different optimum sonication times for better dispersion and higher stability were measured. Here, 0.015, 0.002, 0.05, 0.1% concentration by volume of nanoparticle in water have optimum sonication time 30 min, 45 min, 1 hr 30 min and 2 hr respectively.
2. The dispersion stability of γ-Al2O3 nanoparticles in base fluid was found higher at pH 2.5 to 4.5 correspomdin to zeta potential value 40 mV to 36.1 mV.

List of References

[1] Akoh, H., Tsukasaki, Y., Yatsuya, S., Tasaki, A., 1978, “Magnetic properties of ferromagnetic ultrafine particlesprepared by vacuum evaporation on running oil substrate,” Journal of Crystal Growth, 45, pp. 495–500.

[2] Wagener, M., Murty, B., S., Gu¨nther, B., 1997 “In Nanocrystalline and Nanocomposite Materials II,” edited by Komarnenl, S., Parker, J., C., and Wollenberger H., J., Materials Research Society, Pittsburgh PA, 457, pp. 149–154.

[3] Eastman, J., A., Choi, U., S., S.Li, Thompson, L., J., Lee, S., 1997 “Enhanced thermal conductivity through the development of nanofluids,” Materials Research Society Symposium-Proceedings, Materials research Society, Pittsburgh, PA, USA, Boston, MA, USA, 457, pp.3-11.

[4] Liu, M., Lin, M., C., Tsai, C.,Y., Wang, C., 2006 “ Enhancement of thermal conductivity with cu for nanofluids using chemical reduction method,” Int. J. Heat Mass Transfer, 49 , pp. 3028–3033.

[5] Yu, W., Xie, H., Chen, L., Li, Y., 2010 “Enhancement of thermal conductivity of kerosene-based Fe3O4 nanofluids prepared via phasetransfer method,” Colloids and Surfaces A, 355, pp. 109–113.

[6] Zhu, H., Lin, Y., Yin, Y., 2004 “A novel one-step chemical method for preparation of copper nanofluids,” Journal of Colloid and Interface Science , 277, pp.100–10.

[7] Lee, S., W., Park, S., D., 2012 “ In Cheol Bang, Critical heatflux for CuO nanofluid fabricated by pulsed laser ablation differentiating deposition characteristics,” Int. J. Heat Mass Transf, 55, (23–24), pp. 6908–6915.

[8] Lee, G., Kim, C., K., Lee, M., K., Rhee, C., K., 2011 “Characterization of ethylene glycol based TiO_2

nanofluid prepared by pulsed wire evaporation (pwe) method," Rev.Adv . Mater. Sci., 28, pp. 126-129.
[9] Eastman, J., A., Choi, S., U., S., Li, S., Yu, W., Thompson, L., J., 2011 "Anomalously increased effective thermal conductivities of ethylene glycol-based nanofluids containing copper nanoparticles," Appl. Phys. Lett. 98, 153105 ,10.1063/1.3579522
[10] Lee, S., Choi, S., U., S., Li, S., Eastman, J., A., 1999 "Measuring thermal conductivity of fluids containing oxide nanoparticles," Journal of Heat Transfer, 121, no. 2, pp. 280–289.
[11] Wang, X., Xu, X., Choi, S., U., S., 1999 "Thermal Conductivity of Nanoparticle-Fluid Mixture," Journal of Thermophysics and Heat Transfer, 13, pp. 474–480,
[12] Masuda, H., Ebata, A., Teramae, K., Hishinuma N., 1993 "Netsu Bussei4," 227, pp. 12
[13] Murshed, S., M., S., Leong, K., C., Yang, C., 2005 "Enhanced thermal conductivity of TiO2–water based nanofluid," Int. J. Therm. Sci., 44, pp. 367–373.
[14] Choi, S., U., S., Zhang, Z., G., Yu, W., Lockwood, F.,E., Grulke, E., A., 2001 "Anomalous thermal conductivity enhancement in nanotube suspensions," Appl. Phys. Lett., 79, pp. 2252–2254.
[15] Peng Hu, Wan-Liang Shan, Fei Yu, Ze-Shao Chen, "Thermal conductivity of AlN-ethanol nanofluids", Int. J. Thermophys. 29 (2008) 1968e1973.
[16] Yu, W., Xie, H., Chen, L., Li, Y., 2009 "Investigation of thermal conductivity and viscosity of ethylene glycol based ZnO nanofluid, Thermochim," Acta, 491, pp. 92-96.
[17] Moosavi, M., Goharshadi, E., K., Youssefi, A., 2010 "Fabrication, characterization, and measurement of some physicochemical properties of ZnO nanofluids," International Journal of Heat and Fluid Flow, 31, pp. 599–605.
[18] He, Y., Jin, Y., Chen, H., Ding, Y., Cang, D., Lu, H., 2007 "Heat transfer and flow behaviour of aqueous suspensions of TiO2 nanoparticles (nanofluids) flowing upward through a vertical pipe," Int. J. Heat Mass Transfer , 50, pp. 2272-2281.
[19] Feng, X., Ma, H., Huang S., 2006 "Aqueous-organic phase-transfer of highly stable gold, silver, and platinum nanoparticles and new route for fabrication of gold nanofilms at the oil/water interface and on solid supports," Journal of Physical Chemistry B, 110, 25, pp. 12311–12317.
[20] Yu, W., Xie, H., Chen, L., Li, Y., 2010 "Enhancement of thermal conductivity of kerosene-based Fe3O4 nanofluids prepared via phasetransfer method," Colloids and Surfaces A, 355, pp. 109–113.
[21] Yang, L., Du, K., Zhang, X., S., Cheng, B., 2011 "Preparation and stability of Al2O3 nano-particle suspension of ammonia–water solution," Applied Thermal Engineering , 31, pp. 3643-3647.
[22] Samal, S., Chaira, D., Satpati, B., 2008 "Production and stability of al and cu ultrafine particles in nanofluids" INCOME International Conference On Mechanochemistry And Mechanical Alloying," pp. 80-84.