Investigation of the Influence of Formulation Method on Dispersion Stability and Thermal Properties of Al_2O_3-CC-DW Nanofluids

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Abstract: The present study investigated influence of formulation methods on the physical properties and heat transfer behaviour of aluminium oxide-car coolant-distilled water nanofluids. The nanofluid solutions were formulated via two methods, namely the conventional method (M1), where nanoparticles were added into the CC-DW mixture and the proposed method (M2), where distilled water was added into the Al_2O_3-CC mixture. From the measurement of physical properties, it was observed that the proposed M2 method was more favourable compared to the conventional method because it promoted reductions in density and viscosity values, and also improvement in thermal conductivity. A similar trend was observed when examining nanofluids in the natural convective experiment. The proposed formulation resulted in better dispersion stability when subjected to heat. In addition, the M2 formulation gave higher Grashof (Gr), Rayleigh (R_a) and Nusselt (Nu) numbers. This study demonstrated that dispersion stability, physical properties and thermal performance of nanofluid were remarkably influenced by the preparation process.

Keywords: Nanofluid; Al_2O_3 nanoparticles; Formulation strategy; Natural convection; Sedimentation

دراسة تأثير طريقة التركيب على ثبات التشتت والخواص الحرارية للألانوبدات

Al2O3-CC-DW

الملخص: حققت هذه الدراسة تأثير أساليب الصياغة على الخواص الفيزيائية وسلوك نقل الحرارة من نانوفلويد الماء المقطر المبرد بأكسيد الألومنيوم. وقد صيغت الحلول نانوفلويد عبر طريقتين: هما الطريقة التقليدية (م 1)، حيث أضيفت جسيمات نانوية في الخليط (س س ـ د و) والطريقة المقترحة (م 2)، حيث تمت إضافة الماء المقطر إلى الخليط *Al2O3-CC.* من قياس الخصائص الفيزيائية، لوحظ أن الطريقة (م 2) المقترحة أكثر ملاءمة مقارنة بالطريقة التقليدية لأنها شجعت على تخفيض قيم الكثافة واللزوجة، وأيضا تحسين في الموصلية الحرارية .ولوحظ كذلك اتجاه مماثل عند دراسة نانوفلويد في تجربة الحمل الحراري الطبيعي. الصيغة المقترحة أسفرت عن تحسين استقرار التشتت عند التعرض للحرارة. وبالإضافة إلى ذلك، أعطت الصيغة (م2) عددا أكبر من جراشوف (ج ر)، رايليغ (را)، وأرقام نوسيلت (نو). أظهرت هذه الدراسة أن استقرار التشتت والخصائص الفيزيائية والأداء الحراري للنانوفلويد تأثرت بشكل ملحوظ بعملية التحضير.

1. Introduction

In a modern world of today, the current technology of extended surface and conventional heat transfer fluid or coolant have reached their limitation [1,2]. Therefore, modern engineering system demand a new alternative coolant with superior transport properties. Nanofluids as a promising candidate according to most researcher, need to fulfil the requirements of good long-term stability, less viscous solution and large thermal conductivity value before being acknowledged as a future generation coolant. In response to this issue, researcher tend to chemically modify the solution by either introducing a surfactant or acid/alkali into nanofluid solution. In return, the nanosuspension is stable for months. Nonetheless, the addition of surfactant in return dramatically augment viscosity value and transformed the nanofluid into non-Newtonian behaviour as reported in certain studies [3,4]. Yu et al. [4] found that addition of polyvinyl pyrrolidone into pure ethylene glycol enhanced the viscosity value by about threefold. Jarahnejad et al. [5] reported that viscosity of titanium dioxide nanofluid with trioxadecane acid was approximately 61% higher than that of without surfactant. Ravikumar et al. [6] chemically treated copper-water nanofluid with three

different kind of surfactants, namely PVP, sodium dodecyl sulfate (SDS) and cetyl trimethylammpnium bromide (CTAB). Viscosity measurements showed that nanofluid with PVP surfactant exhibited maximum enhancement of 60%, followed by SDS with 22% increment and 12% by CTAB, respectively. in another studies, addition of surfactant like chitosan, SDS and sodium dodecyl benzene sulfonate (SDBS) to improve stability of multiwalled carbon nanotube in water had transformed the solution into non-Newtonian fluid [7]. Similar observation of non-Newtonian behaviour also observed when the amount of alumina nanoparticles dispersed into car engine coolant greater than 0.4% [3]. While for another kind of zinc oxide-ethylene glycol nanofluid, a non-Newtonian behaviour was observed at nanoparticle concentration of 3.0% and above [4]. In another study, Suganthi and Rajan [8] suggested that modified formulation method of nanofluid also could improved dispersion stability of nanoparticles instead of using surfactant.

For natural convection heat transfer, Wen and Ding [9] reported that addition of titanium dioxide nanoparticles and surfactants (nitric acid and sodium hydroxide) into distilled water reduced the value of natural heat transfer coefficient. Such deterioration was found to be increase with nanoparticles concentration between 0.19% and 0.57%. In a subsequence investigation, similar behaviour also observed by Wen and Ding [10] at high TiO_2 nanoparticles concentration ranging from 0.8% to 2.5%. For alumina-deionized water (Al_2O_3-DI water) nanofluid, Li and Peterson [11] reported that at a given Rayleigh number (*Ra*), deionized water possess the highest Nusselt number (*Nu*) and the lowest values retained by Al_2O_3-DI water nanofluid with maximum volume fraction of 6.0%. Ni et al. [12] also claimed that convective heat transfer coefficient of Al_2O_3-water nanofluid was found to monotonically decrease. Similar trend also observed by Kouloulias et al. [13] although they did not chemically modify the nanofluid. This deterioration was worsening at maximum concentration of 0.12 % when *Ra* value increased, *Nu* value became nearly constant.

Since less attention is given to the formulation method of nanofluid, the present study investigated the influence of modifying formulation method on dispersion stability and physical properties of Al_2O_3-CC-DW nanofluids. Then, heat transfer behaviour of nanofluids that produce through two formulation methods were evaluated in natural convection experiment.

2. Materials and Methods

2.1 Materials

Aluminium oxide (Al_2O_3) nanopowder was commercially purchased from a local supplier. The diameter of primary Al_2O_3 particle provided by manufacturer is 30 nm in spherical shape and its density is 3.7 g/cm3. In the present study, Al_2O_3 nanoparticle was suspended into a base fluid of car coolant and distilled water (CC-DW) with volume ratio of 50:50. The amount of nanopowder suspended in based liquid was corresponding to 0.5 vol. %, and 2.0 vol. %, in which determined as follows:

$$\text{Concentration of nanoparticle (\%)} = \frac{\frac{m_{np}}{\rho_{np}}}{\frac{m_{np}}{\rho_{np}} + \frac{m_{bf}}{\rho_{bf}}} \times 100\% \quad (1)$$

where *m* and ρ are mass and density, while *np* and *bf* denote to nanoparticle and base fluid.

2.2 Formulation of nanofluid

To acquire Al_2O_3-CC-DW nanofluid, two formulation methods were employed to produce the solution as elucidated in Figure 1. In first formulation, represented as Method 1 (M1), which is commonly employed by most researchers, involved suspending a predetermined mass of nanoparticles into a mixture of base liquids. While for the second formulation (M2), at first, a quantity of Al_2O_3 nanoparticles at certain concentration was first suspended in a coolant and then subjected to mixing process by means of shear homogenizer with operating speed of 11 rpm for 25 minutes. At the end of homogenizing process, the required volume of distilled water was added to the Al_2O_3-CC dispersion in order to acquire the final solution of Al_2O_3-CC-DW nanofluid.

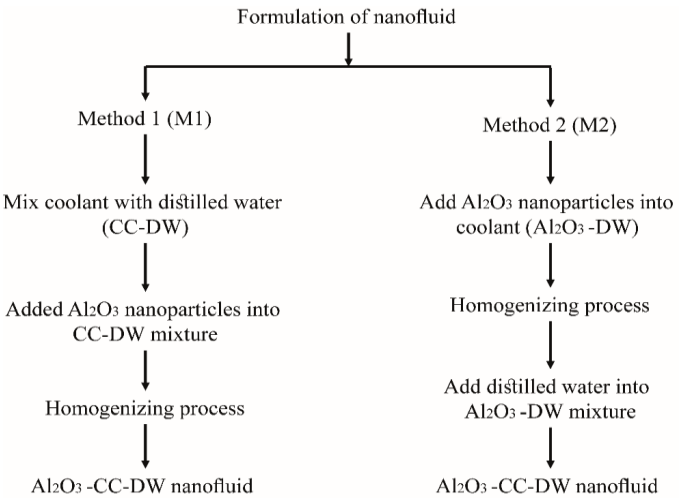

Figure 1. Two types of formulation methods of Al2O3-CC-DW nanofluid.

2.3 Physical properties of nanofluid

Density of Al_2O_3 nanofluid was measured by using hydrometer at room temperature and reported in kg/m^3. Dynamic viscosity was measured using digital rotational viscometer in accordance to ASTM D298 at room temperature. Stability behaviour of Al_2O_3 nanoparticles suspensions within based fluid were evaluated by visual inspection analysis over time. This was done by photographed periodically all samples of Al_2O_3 nanofluid solutions for 84 days.

2.3 Physical properties of nanofluid

A schematic diagram of experimental setup for natural convection heat transfer of Al_2O_3 -CC-DW nanofluids is shown in Figure 2. The experimental setup consists of stainless-steel sample holder, an electrical heater, power supply and K-type thermocouples (TC) connected to a data logger. Four TCs were used to measure temperature of the tested fluid at four different locations as illustrated in top view of test rig in Figure 2b. The location of TCs are 30 mm and 52 mm horizontally and vertically from the centre of test rig. The sample holder was insulated using ceramic fibre to minimize heat losses to the surrounding. In order to maintain constant heat flux boundary condition, the fluid sample was heated by supplying constant DC power to the heater. During the experiment, the sample holder was filled with a constant volume of tested fluid and power of 50 Watts was supplied. The temperature of test fluid was directly measure as a function of time for 3000s.

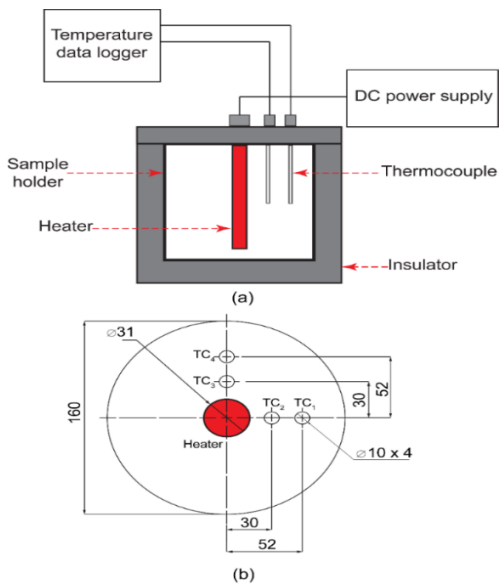

Figure 2. Experimental set up of natural heat transfer experiment.

From the experimental results, thermal conductivity of nanofluid was determined according to the equation:

$$k = \frac{q}{4\,\pi\,(T_2 - T_{1)}} \ln\left(\frac{t_2}{t_1}\right) \tag{2}$$

where T_1 and T_2 are temperature at different time of t_1 and t_2, while q is power input to the system, which was assumed constant during natural convective experiment. Moreover, specific heat of the fluid was calculated using the expression of

$$C_p = \frac{Q}{m\,\Delta T} \tag{3}$$

where m is mass of nanofluid, *Q* is flow of heat and ΔT is temperature difference between two time. Next, Grashoff number, *Gr* was determined as

$$Gr = \frac{gB(T_s - T_\infty)L^3}{\nu^2} \tag{4}$$

where L is a characteristic length of heat source, ν is kinematic viscosity, B is thermal expansion coefficient and both Ts and T∞ are defined as instantaneous temperature. For nanofluid solution, thermal expansion coefficient of nanofluid is calculated based on the following formula [15]:

$$B_{nf} = \frac{\phi\,\rho_{np}\,B_{np} + (1-\phi)\left(\rho_{bf}\,B_{bf}\right)}{\rho_{nf}} \tag{5}$$

where subscripts of np, bf and nf are refer to nanoparticle, base fluid and nanofluid and ρ is density value of the constituents. Thermal expansion coefficient of Al_2O_3 nanoparticle was taken to be 8.46×10^{-6} K^{-1}, while for distilled water the value varies between 1.5×10^{-4} K^{-1} and 6.2×10^{-4} K^{-1}

for temperature ranging from 15ºC to 80ºC [16]. Lastly, thermal performance of nanofluid was evaluated based on Nusselt number which is determined according to equation 6 [17]:

$$Nu = \left\{ 0.60 + \frac{0.387Ra^{\frac{1}{6}}}{\left[1 + \left(\frac{0.559}{Pr}\right)^{\frac{9}{16}}\right]^{8/27}} \right\}^2 \quad (6)$$

where *Ra* and *Pr* are Rayleigh number and Prandtl number, where *Pr* number is expressed as $Pr = \frac{C_p}{k}$ and *Ra* = *Gr* x *Pr*. After that, thermal properties of the solution such as thermal conductivity, specific heat, Grashoff number, Rayleigh number and Nusselt number were investigated by plotting those properties as a function of experimental period.

3. Results and Discussions

3.1 Physical properties of nanofluid

Figure 3 and Figure 4 presented density and viscosity of nanofluids. It can be seen that density of the Al_2O_3 -CC-DW nanofluids were conspicuously higher than that of their base fluids, and the augmentation in density value was linear with volume concentration. And this result was consistent with correlation proposed by Pak and Cho [18] as given below:

$$\rho_{nf} = \phi\rho_p + (1-\phi)\rho_{bf} \quad (7)$$

where ρ_{nf} is nanofluid density, ρ_{bf} is fluid density, ρ_p is particles density and ϕ is volume concentration of nanoparticles. The density of nanofluid produced via the M2 formulation was consistently lower than that of the nanofluid formulated using M1. At 5.0 vol.% nanoparticle concentration, the difference of density between formulation M1 and M2 is 0.18% and 4.0% for M2, respectively. This observation demonstrated that the density was strongly dependent on the

formulation method, in which the chemical interactions between the constituents had a dramatic influence on the density of nanofluid.

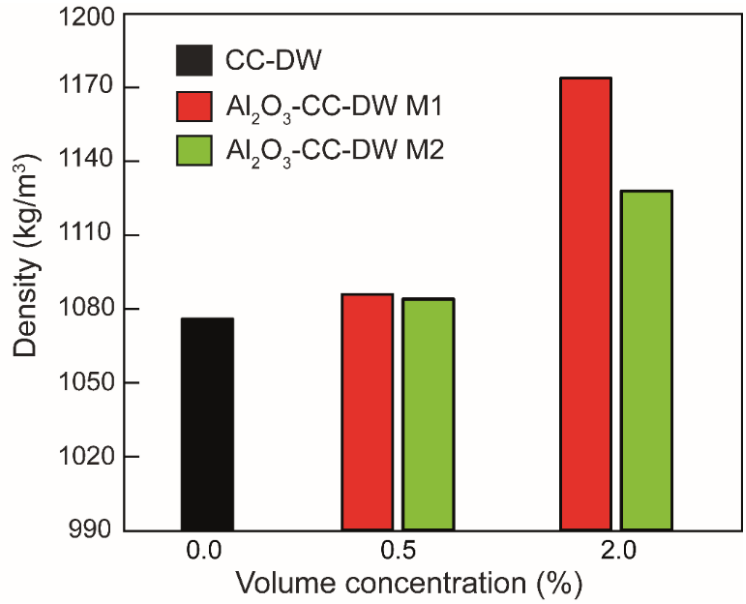

Figure 3. Effect of nanofluid's formulation and concentration on the density.

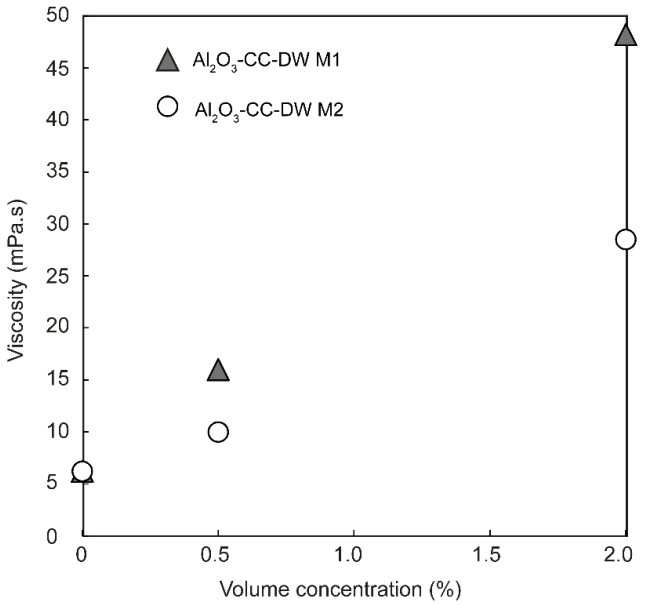

Figure 4. Viscosity of Al_2O_3-CC-DW nanofluid with 0.5 and 2.0 wt%.

As can be seen in Figure 4, viscosity of nanofluids were substantially larger than that of its base fluid, a mixture of car coolant with distilled water (CC-DW). This was because the suspended solid particles triggered a change in the velocity profile of base fluid by creating a secondary velocity profile around them and consequently a higher shear stress was generated at solid-liquid interface [14]. Then, the higher shear stress that evolved in a nanofluid system would augment the resistance of the liquid to motion. It was also observed that viscosity of nanofluids enhanced linearly with Al_2O_3 concentration. As the amount of Al_2O_3 nanoparticles suspended in base liquid escalated from 0.5% to 2.0%, more shear stress was generated at the solid-liquid interface, and this induced more alteration in the velocity profile of base fluid.

Besides, Al_2O_3-CC-DW nanofluids formulated by M2 were apparently lower than that of M1. According to [8,19], the addition of distilled water into Al_2O_3-CC mixture, as in the formulation M2, led to the disruption of the intermolecular hydrogen bonds in the coolant by water molecules, as elucidated in Figure 5b. Any reordering or disrupting of the hydrogen bonding in ethylene glycol molecules would result in a reduction of viscosity of the bulk solution, as reported in a previous experimental research [8,20]. Suganthi and Rajan [8] reported that the addition of water into zinc oxide-proplyene glycol (ZnO-PG) mixture to acquire ZnO-PG-water nanofluid, led to reduction in viscosity of about 11.4% at 2.0 vol. %. In another study, Christensen et al. [20] studied the effect of suspended iron oxide nanoparticle into various kind of solvents such as ketone, glycerol and water. Among the solvents, only water exhibited a good dispersion due to hydrogen bonding between water molecules. The authors claimed that the strong bonding not only promoted good stability behaviour but also induced the nanofluid became less viscous.

Stability of nanofluids formulated through Method 1 (M1) and Method 2 (M2) are shown in Figure 6 and Figure 7. For the formulation M1, nanofluid with 0.5 vol% was visually stable at day 0 only, while nanofluid with 2.0 vol.% managed to stable up to 3rd day. Meanwhile, for the solution formulated by M2, nanofluids of both concentrations were observed to be stable without any sediment layer at day 0 only. The results conspicuously indicated that sedimentation rate of M1 Al_2O_3 nanofluids was substantially slower than the M2 nanofluids. This observation contradicted with the findings reported by Suganthi and Rajan [8]. The researcher inferred that the addition of distilled water into nanoparticle-ethylene glycol (EG) dispersion could improve the stability behaviour as direct contact between water molecules and the nanoparticles had been minimized and this could reduce the aggregation rate and sedimentation rate eventually [8,20].

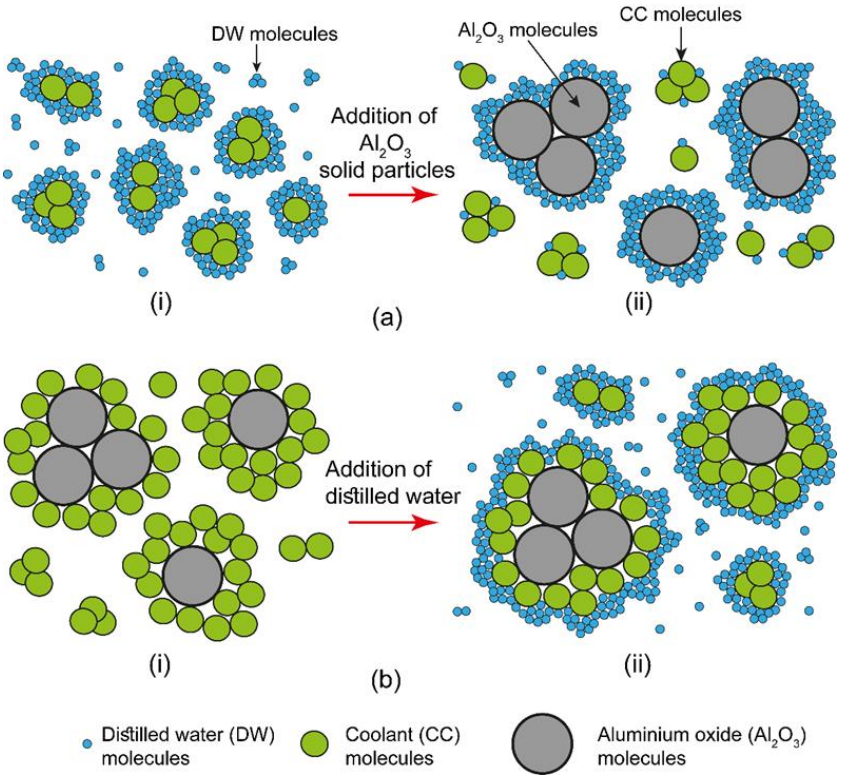

Figure 5. Mechanism of liquid layer in nanofluid when formulated with a) Method 1 and b) Method 2 (ai: CC-DW mixture, aii: Al_2O_3-CC-DW-M1 nanofluid, bi: Al_2O_3-CC mixture, bii: Al_2O_3-CC-DW-M2 nanofluid).

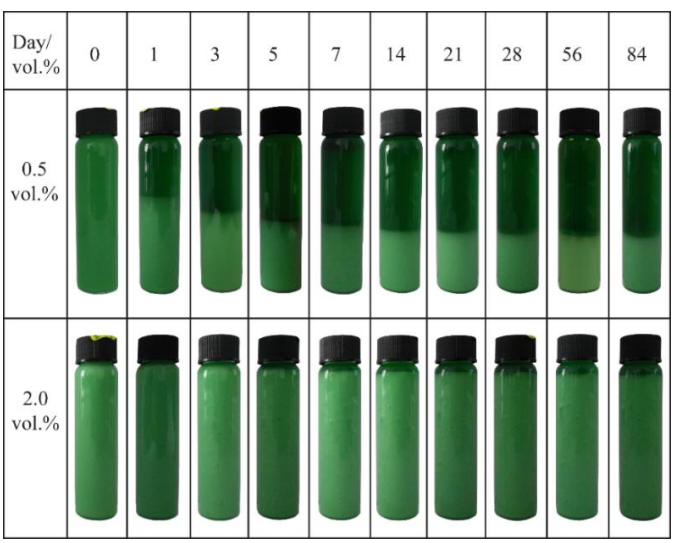

Figure 6. Stability of nanofluid formulated by method 1 as a function of time

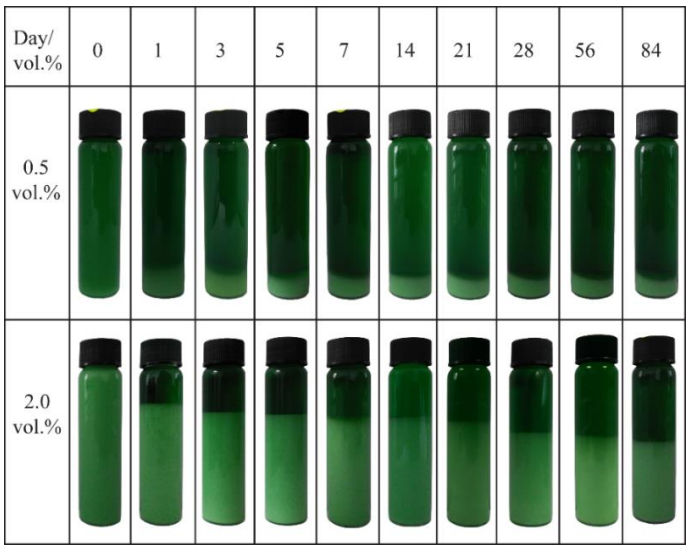

Figure 7. Stability of nanofluid formulated by method 2 over time.

The contradictory result might be attributed to the strong intermolecular forces due to the change of flow behaviour from Newtonian to non-Newtonian characteristic. As found in Figure 4, Al_2O_3-CC-DW nanofluids formulated by M1 were highly viscous especially at high concentration of 2.0% and at the same time demonstrated excellent stability behaviour. These results were comparable with observations reported by Kole and Dey [3]. Their rheological studies demonstrated greater than that 0.4%, nanofluid behave like non-Newtonian fluid. They found that nanofluid that demonstrated a non-Newtonian characteristic exhibited a very good stability behaviour for over 80 days in comparison to the nanofluid that demonstrated the Newtonian fluid. Hence, it was strongly believed that the good stability behaviour of the M1 nanofluid compared to that of M2 was due to the non-Newtonian property.

Apart from that, in both formulations, it was found that Al_2O_3 -CC-DW nanofluid with 0.5 vol.% exhibited the fastest settling rate compare to that of 2.0 vol.%. This result contradicted with the previous observation reported by [21,31]. The authors claimed that the stability of the Al_2O_3-DW nanofluid became worse at high concentration of nanoparticles because when the amount of solid particles increased, the distance between each particles reduced. Hence, there was a higher chance of collisions to occur between the particles, which would trigger rapid aggregation process. However, the interpretation was different from Amrollahi et al. [22] who investigated the effect of sonication time on the settling rate of carbon nanofluids. They observed that at a shorter sonication time less than 10 hours, nanofluid with 2.5 vol. % settled faster than nanofluid with 0.5 vol.%

concentration. Meanwhile, at a longer sonication time of 25 hours, 2.5 vol. % carbon nanofluid was physically stable than the 0.5 vol. % solution. Amrollahi and his co-worker concluded that in order to produce a homogenous and stable suspension, different volume concentrations of nanofluid require different preparation times. Moreover, a prolonged preparation time caused the solution to be unstable due to the excessive energy delivered to the fluid system [23-25]. Hence, the faster sedimentation rate of 0.5% Al_2O_3 -CC-DW nanofluid was because of the excessive mixing energy received by the solution. When this happens, nanoparticles are encouraged to re-agglomerate more rapidly and subsequently increased the sedimentation rate.

3.2 Thermal Conductivity and Specific Heat Capacity of nanofluid

The thermal conductivity (k) of Al_2O_3–CC–DW nanofluids is investigated with different formulation methods and volume concentration as presented in Figure 8. It was conspicuous from the figure that k value of nanofluids were relatively larger than that of their base fluid and the trend increased linearly with concentration of nanoparticle. It was also noticeable that k value of nanofluids with formulation M2 were larger in comparison with conventional method of M1. Therefore, the addition of water into the CC-DW mixture not only reduced the viscosity as in Figure 4 but also enhanced the thermal conductivity property of nanofluid.

Figure 9 presents the results of specific heat capacity (C_p) Al_2O_3–CC–DW nanofluids and its base fluids against experimental time between 500s and 3000s. It was seen that specific heat value of 0.5% and 2.0% Al_2O_3–CC–DW nanofluids were increased with heating period. These results exhibited that capability of all solutions to absorb heat enhanced with increasing experimental time along with temperature rise.

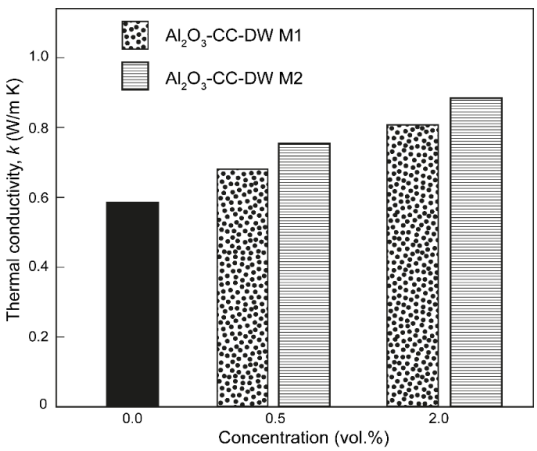

Figure 8. Thermal conductivity of Al_2O_3–CC–DW nanofluids.

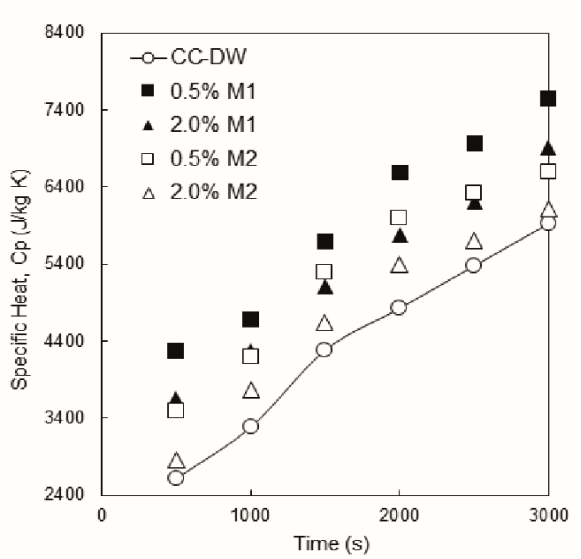

Figure 9. Specific heat capacity of Al_2O_3 –CC–DW nanofluids.

Apart from that, it was found that C_p value of fluid containing Al_2O_3 nanoparticles was always larger than that of CC–DW base fluid. However, C_p value experienced a reduction in the enhancement when further increased volume concentration of nanoparticles from 0.5% to 2.0%. Such behaviour of C_p over nanoparticle concentration had been observed in other kinds of nanofluid [26,27]. Ijam et al. [26] found that the C_p value of graphene oxide–deionized water–ethylene glycol nanofluid substantially increased with nanoparticle concentration from 0.01 to 0.05 wt.%, but the value decreased when the concentration exceeded 0.07%. Similarly, for multiwall carbon nanotube–water– ethylene glycol nanofluid, Kumaresan and Velraj [27] reported that the reduction in the improvement of C_p occurred at higher concentration of beyond 0.45%. These experimental observations indicated that there was a limit in the amount of nanoparticle suspended in the base fluid, where it become a margin between the enhancement and reduction trend of specific heat capacity.

3.3 Thermal Performance: Grashof number, Rayleigh number and Nusselt number

From the natural convective experiment, thermal performance of Al_2O_3–CC–DW nanofluid is evaluated in term of Grashof number (*Gr*), Rayleigh number (R_a) and Nusselt number (*Nu*) as depicted in Figure 10, Figure 11 and Figure 12, respectively. From Figure 10, it was found that only the base fluids of CC-DW mixture experienced a sharp augmentation of *Gr* value, while all nanofluids behaved contrarily against heating period. Besides that, *Gr* value of nanofluids were lowered than that of base fluid and deteriorated with increasing of nanoparticle concentration.

These results demonstrated that the presence of nanoparticles in the fluid medium retarded the motion of fluid and energy exchange rate driven by buoyancy force.

For nanofluid system, the existence of nanoparticle was expected to augment the temperature gradient and subsequently velocity of fluid flow because the Brownian motion of solid particles became aggressive with increasing temperature. However, from the experimental results, it was observed that the presence of nanoparticles decelerated the velocity of fluid across the enclosure as the *Gr* values were lower than that of base fluid. These findings were absolutely contradicted with the results reported by [15,28,30] as the respective authors inferred that by suspending nanoparticles into base fluid, the fluid flow and velocity distribution were explicitly enhanced, thus leading to the escalation in the rate of energy transfer.

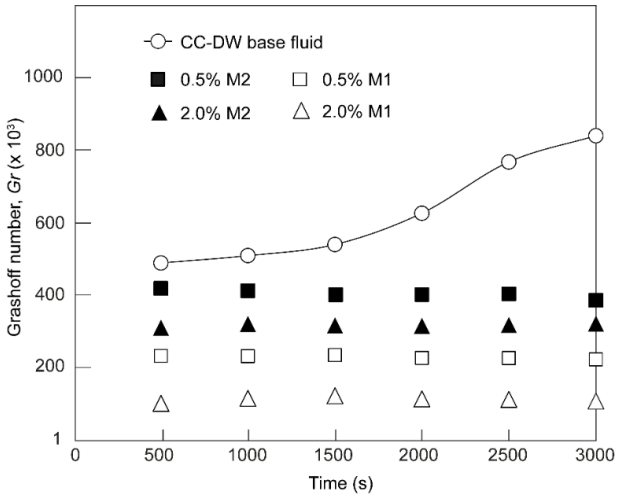

Figure 10. Grashoff number of CC-DW and Al_2O_3 nanofluid of formulation M1 and M2.

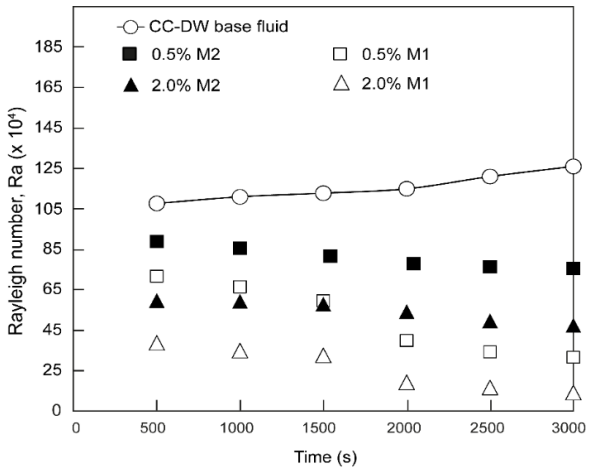

Figure 11. Reduction of Rayleigh number of Al_2O_3–CC–DW nanofluids over time.

A similar trend of *Gr* of nanofluid as a function of heating was observed for the Rayleigh number (R_a) as presented in Figure 11, where only the base fluid exhibited an increment in R_a value and degradation of R_a of nanofluids became worse with increasing nanoparticle concentration. This result was identical with the numerical study done by Snoussi et al. [29]. The authors suggested that at high nanoparticle concentration, the solution became highly viscous and tend to retard the fluid movement and consequently the convection effect. They inferred that R_a behaviour was very sensitive towards viscosity of nanofluid. For nanofluid, the degradation effect of both *Gr* and R_a became less when formulated with M2 compared to M1. It might be attributed to the enhanced physical properties of nanofluid as a result of the strong liquid layering of hydrogen bonding in formulation M2.

From the *Gr* and R_a graphs, the presence of nanoparticle deteriorated the *Nu* as shown in Figure 12, where *Nu* value of nanofluids were profoundly worsen with increasing of nanoparticle concentration. Experimental investigation done by Kouloulias et al. [13] also demonstrated similar trend, where the suspension of alumina nanoparticle in deionized water had significantly degraded the heat transfer coefficient and the values degenerated with increasing concentration of alumina. The authors claimed that the deterioration of heat transfer performance was attributed to the severe sedimentation of nanoparticles when subjected to the heat as they discovered the deposition of Al_2O_3 nanoparticles on the hot plate though the nanofluid was in a good stability condition before the experiment started.

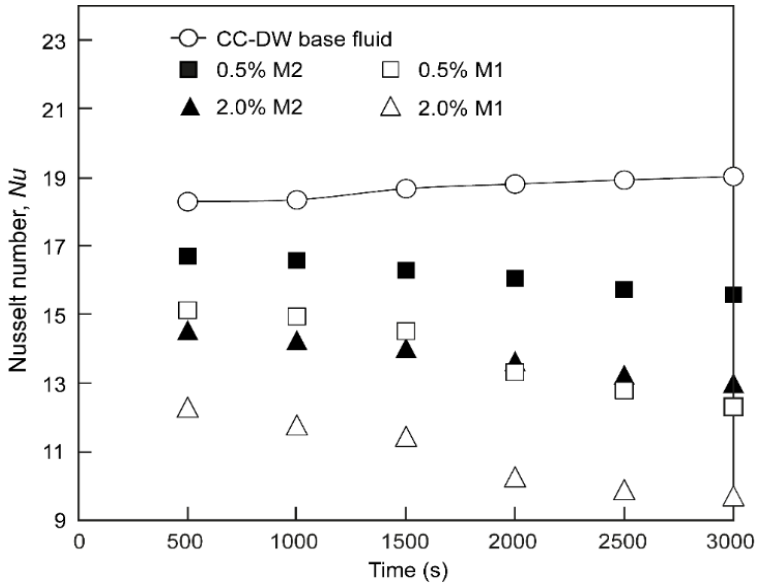

Figure 12. Nusselt number of Al_2O_3–CC–DW nanofluids as a function of time.

To prove such occurrence was also present in the present study, a dummy heating test was conducted as shown in Figure 13 since sedimentation phenomenon cannot directly observed during natural convective experiment. It was done by heating the nanofluid at a constant power input for 30 minutes. After 30 minutes of heating, nanofluid with 0.5 and 2.0 vol.% were conspicuously separated into two layers, where the lower layer which closed to the heat source is dominated by the white layer that was believed belong to Al_2O_3 nanoparticle. These results obviously showed that imposing heat to the nanofluid would rapidly augment the rate of aggregation process, induced the particles to free themselves from the bulk fluid and eventually deteriorated heat transfer performance of the solution.

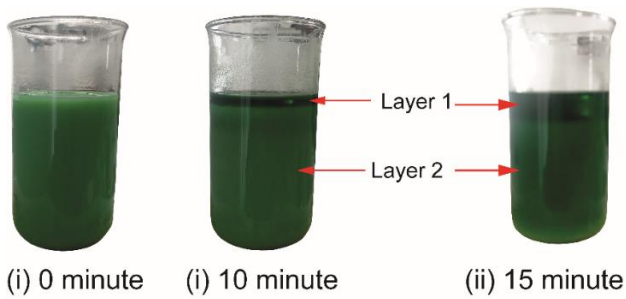

Figure 13. Separation of Al_2O_3–CC–DW nanofluids solution when subjected to heat.

4. Conclusion

In the present study, Al_2O_3 nanoparticles are dispersed in the mixture of car coolant and distilled water (CC-DW). Modifying formulation strategy has been utilized in order to acquire Al_2O_3-CC-DW nanofluid with good stability behaviour. The results revealed that liquid layering of molecule has a huge influenced on the stability behaviour, density, viscosity, thermal conductivity and specific heat capacity properties of nanofluid. Enhancement in physical properties were observed in Al_2O_3 nanofluid with new formulation of M2. From the natural convection experiment, it was found that the addition of Al_2O_3 nanoparticles in CC-DW base fluid has deteriorated the Grashof number, Rayleigh number and Nusselt number. Deposition of nanoparticles when subjected to heat was the main contributory factor of this deterioration. However, the deterioration effect became less when employing nanofluid with new formulation.

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