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An experimental study on thermal characteristics of nanofluid with graphene and multi-wall carbon nanotubes

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Abstract: High-thermal conductivity enhancement of nanofluid is one of the promising topics of the nanoscience research field. This work reports the experimental study on the preparation of graphene (GN) and multi-walled carbon nanotubes (MWCNTs) based nanofluids with the assistance of sodium dodecyl benzene sulfonate (SDBS) and sodium dodecyl sulfate (SDS) surfactants, and their thermal behaviors. The present work suggests not a solution, but a solution approach and deduces a new conclusion by trying to resolve the agglomeration problem and improve the dispersibility of nanoparticles in the base fluid. The analysis results of FESEM, thermal conductivity, diffusivity, effusivity and heat transfer coefficient enhancement ratio of nanofluid with surfactants SDS and SDBS expose strong evidence of the dispersing effect of surfactant on the making of nanofluid.

Key words: graphene; carbon nanotube; surfactant; dispersibility; thermal conductivity

1 Introduction

The researches of recent decades have demonstrated the advantages of nanofluids compared with those fluids containing millimeter or micrometer size particles [1]. Nanofluid with a high thermal conductivity enhancement may be potentially applicable in heat sink applications such as coolant [2]. The concept of nanofluid was first introduced by CHOI and EASTMAN [3] who quantitatively analyzed some potential benefits of nanofluids [4]. Within a few years, many research projects [1, 5] were conducted in order to produce more stable suspensions with well dispersion, as well as homogenization. Some surfactants, such as sodium dodecyl sulfate (SDS), sodium dodecyl benzene sulfonate (SDBS), are under experiment for using as dispersant for these nanofluids [6] as well as PCA in the grinding process [7]. Ultrasonication is used for dispersing nanoparticles in a base fluid like distilled water, whereas, grinding method is applied for increasing specific surface area [8] which helps to decrease the sphericity of particles and as a result of this, thermal conductivity increases [4]. Transparent conductive films (TCFs), which were widely used in transistors [9] as well as in solar cells, are fabricated by indium tin oxide [10]. But the rising cost of indium, high temperature processing in production and brittleness of ITO [11−12] have introduced some emerging alternatives, such as CNTs (carbon nanotubes) [13], GN (graphene) [14], metal or metal nanowires [15] and hybrids of these [16].

Graphene (GN) has two-dimensional extended honeycomb network of sp^2 hybridized carbon atoms [17], high electron mobility [18], excellent mechanical and chemical properties [19], high thermal conductivity (5000 W/(m·K)) [20] and twice particular surface area compared with SWCNTs [11], whereas CNTs, first made in the early 1990s [21], carry some features such as one of the lightest [22], strongest [23], stiffest [23], electrically conductive [24−25] nanoparticles with thermal conductivity of 3500 W/(m·K) [26]. Moreover, individual CNTs have ON/OFF current ratios higher than

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10^5 [27] and carry three orders of magnitude higher current than copper [28]. The fundamental difference between GN and CNTs is that optically, GN is reflective with little absorption while CNT is the darkest material with significant optical absorption [27]. However, GN and CNTs share very similar carbon chemistry and, therefore, the methods and understanding for CNTs can be transferred to GN research [27]. The intrinsic tendency to agglomerate [29] in water due to the hydrophobic surface [8] is the chief obstacle to preparing GN and CNTs nanoparticles based nanofluids that are under research with a significant challenge. This work tried to solve this problem and chose wet grinding and ultrasonication in order to minimize this problem while making nanofluid. The reason for wet grinding is to decrease the sticking problem and the possibility of decomposition of surfactants that produces contamination. Surfactant controls the welding and fracturing [7] while grinding process and during ultrasonication, and it is used as dispersant [1]. So, in order to get the both advantages, surfactants were added with nanoparticles from starting of the process. This work does not suggest a solution but a solution approach and tries to deduce a new conclusion by giving evidence by showing the effect of surfactant SDBS on wet grinding process and SDS and SDBS on sedimentation, and thermal conductivity, as well as analyzing effusivity, diffusivity and heat transfer coefficient enhancement ratio precisely for better decision making. Here, it should be noted that the results of this work are only for comparison and do not indicate the optimum conditions for this process.

2 Experimental setup

2.1 Materials

Raw MWCNTs with a ~20 nm diameter, ~5 μm length (Carbon Nanomaterial Technology Co., Ltd, South Korea) and graphene nanopowder with 8 nm (average flake thickness) flakes, average particle size ~550 nm, specific surface area 100 m^2/g, and 99.9% purity (graphene supermarket) were used in this experimental study.

Sodium dodecyl benzene sulfonate (SDBS, $C_{18}H_{29}NaO_3S$) with hard type, 348.48 relative molecular mass (Tokyo Chemical Industry Co., Ltd) and sodium dodecyl sulfate (SDS, $CH_3(CH_2)_{11}OSO_3Na$) with 288.38 relative molecular mass (Junsei Chemical Co., Ltd) surfactants were used as dispersant as well as controller of wielding and fracturing during grinding process. Distilled water (DW) was used as a base fluid for making nanofluids.

2.2 Grinding process

Grinding is useful for increasing the specific surface area and upgrading the dispersibility of nanoparticles in a base fluid. A planetary ball mill machine (HPM-700) (Haji Engineering, Korea) is used to shorten the length of the nanoparticles of GN and MWCNTs by both dry and wet grindings. Mono-sized (3.0 mm) spherical zirconia (ZrO_2) balls were used as the collision medium. The agitator-applied rotation speed was 500 r/min, and the grinding time was 1 h for each grinding process. During the grinding process, nanoparticles were subjected to high energy inter-particle and milling ball collisions. Thus, particle breakage occurred due to the relative impact velocity of the ball colliding with each other or against the mill, pot and wall while grinding. The details of the grinding process were described elsewhere [30]. The grinding process was done with and without adding surfactants where surfactants were used as process control agent [7]. Both surfactants (SDBS and SDS) were added individually while wet grinding. Surfactants were added only in wet grinding process in order to avoid contamination as a consequence of the decomposition of these organic surfactants [7].

At first, dry grinding was performed for both GN and MWCNTs without surfactant in order to observe the agglomeration problem. Then, wet grinding was performed for the same nanoparticles with the same amount. Finally, wet grinding with adding the surfactants was performed to observe the effect of the surfactant in balancing welding and fracturing processes while shortening the particle size. Nanoparticles and surfactants were hands mixed. The mass ratios between nanoparticle and surfactant were 1/3, 3/1, 1/1, 1/2 and 2/1 while adding the surfactant before grinding process.

2.3 FESEM

Morphological analysis of raw and ground nanoparticles of both MWCNTs and GN was studied by field emission scanning electron microscope (FESEM) (JSM-6710F, JEOL). Nanoparticles of MWCNTs and GN were ground at 500 r/min for l h in wet condition in the presence of SDBS. The total grinding process was described in section 2.2. After wet grinding, some of these nanoparticles were dried at 90 °C for 30 min in furnace in order to analyze the morphology by FESEM.

2.4 Nanofluid

Dispersing nanoparticles in aqueous media with the use of surfactant is one of the three major approaches to make nanofluid by ultrasonication [31]. In this work, Branson ultrasonic cleaner model 1501E-DTH (Branson Ultrasonic Corporation 41, Danbury, CT 06813, USA) was used to disperse ground GN and MWCNTs in distilled water used as a base fluid. During ultrasonication, extra surfactant was not added since it was added while grinding. Ultrasonication served to impart enough energy to separate the nanoparticles from each other long enough for surfactant to surround the nanoparticles and prevent them from agglomerating [32]. Since excessive sonication causes defect in CNTs [33], the ultrasonication time for both GN and MWCNTs was 20 min. Later, 40 min ultrasonication was performed for GN-SDS nanofluid. Nanofluids were categorized into two groups: one was for sedimentation test where mass ratios (nawoparticles/surfactant)) were 1/3, 3/1, 1/1, 1/2 and 2/1 and the another one was for thermal conductivity measurement and analysis where mass ratios were 1/1 and 2/1.

2.5 Testing apparatus

In order to get continuous determination of the thermal conductivity as well as other necessary information such density, specific heat capacity for calculating thermal diffusivity, thermal effusivity as well as heat transfer coefficient enhancement ratio, the LAMBDA system measuring instrument was used. Precisely calculation of the thermal conductivity by a theoretical approach is almost impossible [4], so experimental approach is mandatory. Moreover, for this, the in-stationary transient hot-wire method, i.e. LAMBDA system is used. At first, hot wire apparatus was calibrated by measuring the thermal conductivity of distilled water (DW) which has known thermal conductivity. The difference between the standard value and the experimental value is shown in Fig. 1. Moreover, the error is within 1.5% with respect to the standard value.

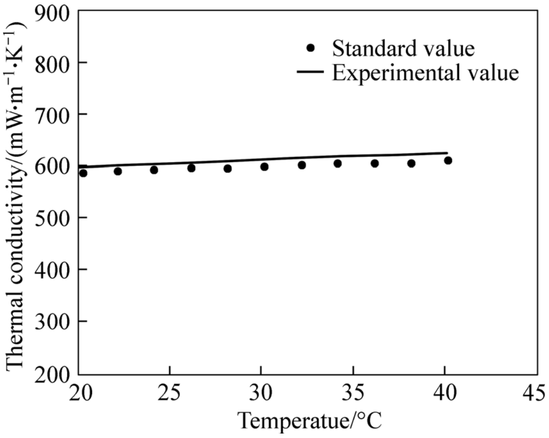

Fig. 1 Graph of standard value and experimental value of thermal conductivity of distilled water

3 Results and discussion

3.1 Result of grinding

As shown in Fig. 2, both GN and MWCNTs nanoparticles agglomerated and adhered to vials and balls. More adherence to grinding media causes less amount of final ground nanoparticles. This severe

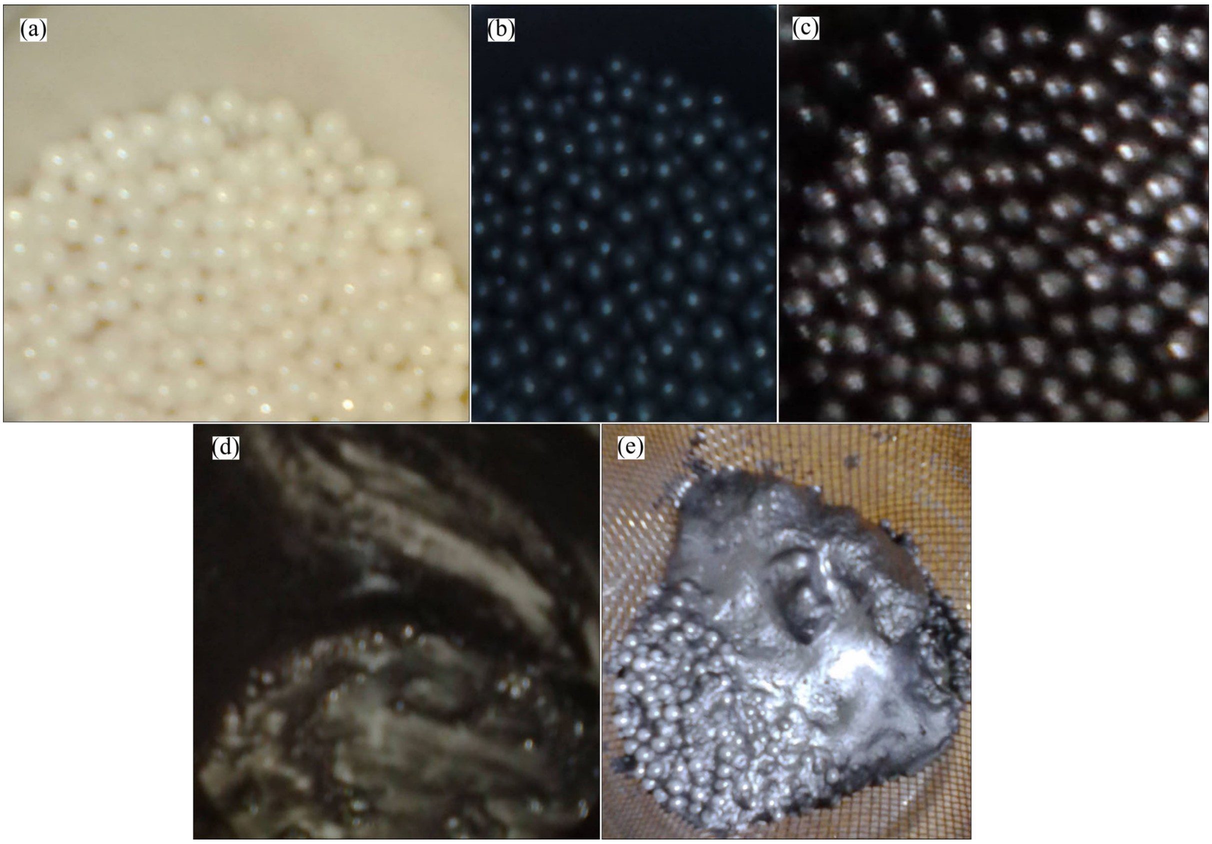

Fig. 2 Grinding container and balls: (a) Before grinding; (b) Dry grinding of MWCNTs nanoparticles; (c) Dry grinding of GN nanoparticles; (d) SDBS based wet grinding of MWCNTs nanoparticles; (e) SDBS based wet grinding of GN nanoparticles

adherence problem occurred while dry grinding was performed (Figs. 2(b) and (c)). However, wet grinding slight reduced this adherence problem. During wet grinding, less adherence of MWCNTs to grinding media occurred than that of GN, and it was obvious that only wet grinding was not enough to minimize this problem. Modifying of the deforming nanoparticles by introducing suitable organic material, i.e. surfactant, is one of the two techniques to hinder the clean metal to metal contact [34]. In this regard, SDBS and SDS were introduced in the grinding process as a process control agent. Only, SDBS based wet grinding images of MWCNTs and GN are presented in Figs. 2(d) and (e), respectively. While collecting the ground nanoparticles, there was foam for both surfactants that hindered the accurately collecting of the grounding nanoparticles. As shown in Figs. 2(d) and (e), there was no agglomeration problem, and the mass ratio of nanoparticles and surfactant was 1/1.

Figures 3 and 4 show the raw nanoparticles of MWCNTs and GN and their wet grinding particles at 500 r/min for 1 h in the presence of SDBS surfactant, respectively. The mass ratio of nanoparticles to surfactant was 1/1. As shown in Fig. 4, nanoparticle size decreased significantly. It is evident that the entanglement of raw MWCNTs decreased, which increased the straightness ratio. After grinding, specific surface area for both GN and MWCNTs nanoparticles increased.

3.2 Sedimentation of nanofluids

WUSIMAN et al [1] observed that SDBS is the better dispersant for MWCNTs with particular mass ratio in order to disperse properly in distilled water than SDS. For this reason, in this work, only nanofluids of GN by adding SDBS and SDS individually were made. The mass ratios were 1/3, 3/1, 1/1, 1/2 and 2/1 (as received from wet grinding). Sedimentation method is the simplest way to evaluate the stability of nanofluids where sediment mass or the sediment volume of nanoparticles in a nanofluid under an external force field is an indication of the stability of the characterized nanofluid [35]. As shown in Fig. 5, five GN−SDBS nanofluids with different mass ratios were made in order to find out the effective mass ratio of GN to SDBS for better dispersibility. After four days, sedimentation was observed as shown in Fig. 6. It was obvious that only less amount of SDBS (3/1 mass ratio) was suitable for better dispersibility (sample e). It is also obvious that SDBS was not suitable for making GN nanofluids at all. In addition, GN−SDS nanofluids were also made as shown

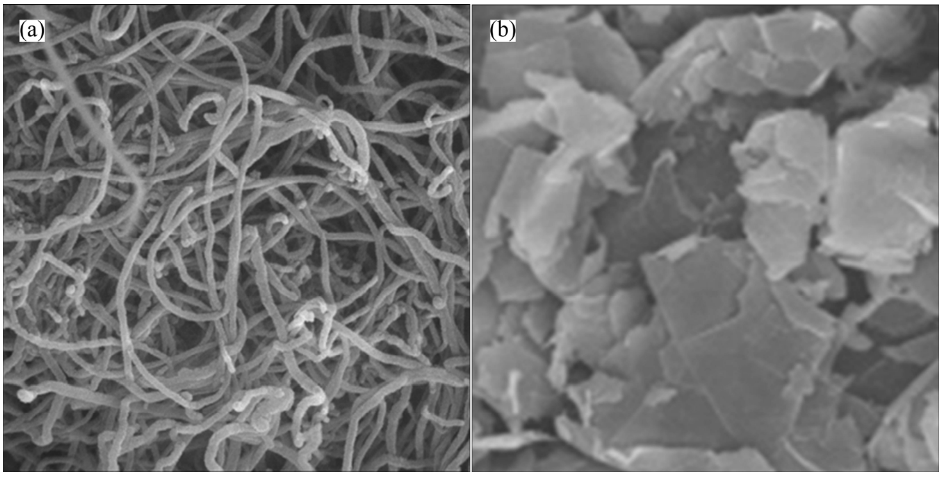

Fig. 3 FESEM patterns of raw MWCNTs (a) and raw GN (b)

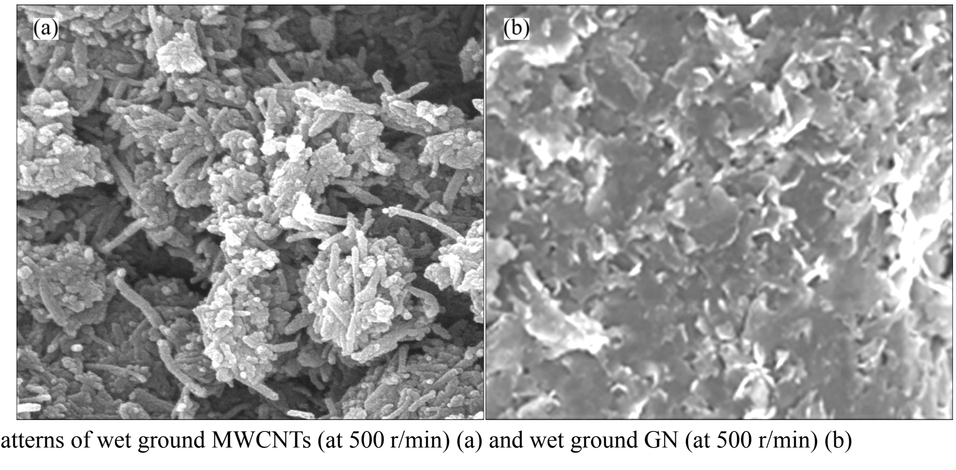

Fig. 4 FESEM patterns of wet ground MWCNTs (at 500 r/min) (a) and wet ground GN (at 500 r/min) (b)

in Fig. 7. Like GN−SDBS nanofluids, lower mass ratio showed better dispersibility than those of other four nanofluids. However, this dispersibility was better than that of GN-SDBS as shown in Fig. 8. ISLAM et al [36] found that graphene required more planar surfactant for dispersion due to its two-dimensional nature. The relative moleculer mass of SDS is 288.38 g/mol, whereas 348.48 g/mol is for SDBS. Moreover, the chemical structure of SDS is more planar than SDBS. As a consequence, GN−SDS nanofluid showed better dispersibility.

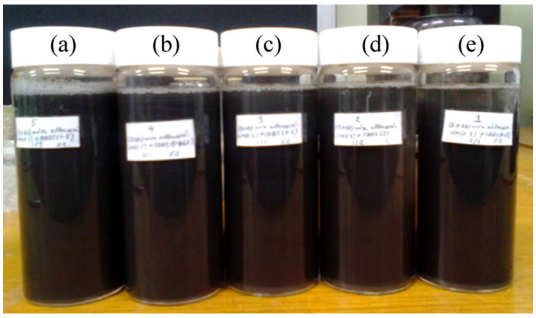

Fig. 5 Ultrasonication of GN and SDBS with different mass ratios of 1/3 (a), 3/1 (b), 1/1 (c), 1/2 (d) and 2/1 (e)

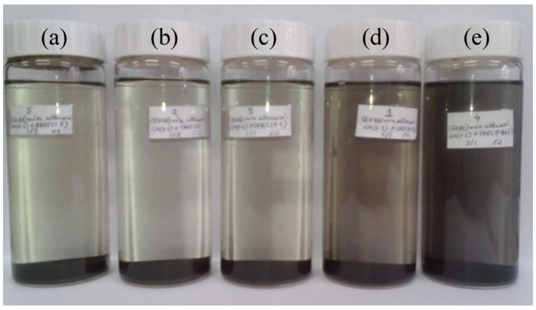

Fig. 6 After four-day settlement of ultrasonication of GN and SDBS with different mass ratios of 1/3 (a), 1/2 (b), 1/1 (c), 2/1 (d) and 3/1 (e)

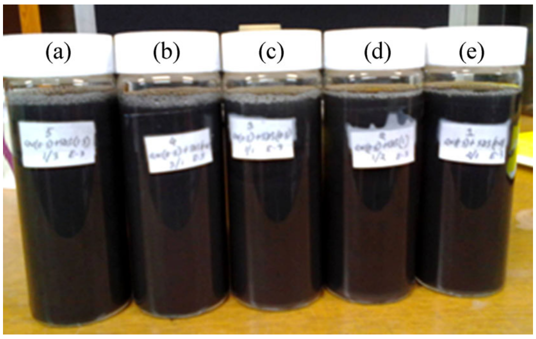

Fig. 7 Ultrasonication of GN and SDS with different mass ratios of 1/3 (a), 3/1 (b), 1/1 (c), 1/2 (d) and 2/1 (e)

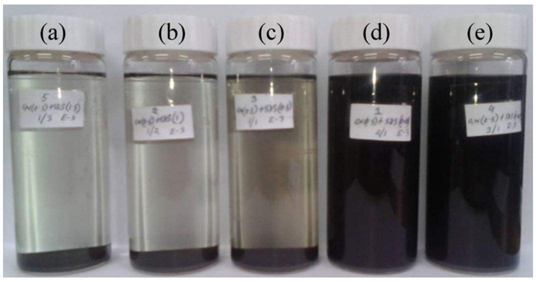

Fig. 8 After four-day settlement of ultrasonication of GN and SDS with different mass ratios of 1/3 (a), 1/2 (b), 1/1 (c), 2/1 (d) and 3/1 (e)

3.3 Thermal conductivity

The effect of size of nanoparticles on the thermal conductivity can be explained from the two aspects: the particles increase the thermal conductivity of nanofluid and the chaotic movement of the particles strengthens energy transport process [4]. The thermal conductivity which is the property of a material to conduct heat measured by LAMBDA system and the related graph has been shown in Fig. 9.

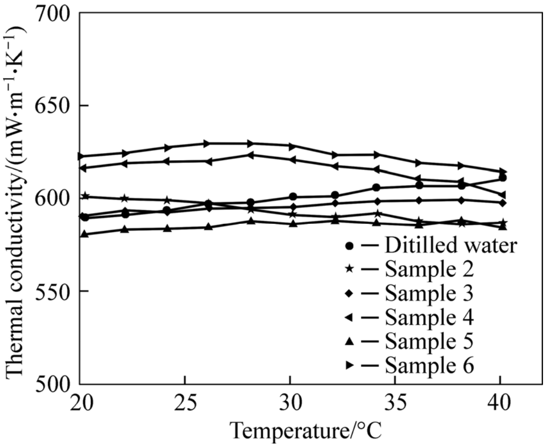

Fig. 9 Thermal conductivity vs temperature curves (Sample 1: distilled water; Sample 2: GN−SDBS (1/1) nanofluid, wet grinding, 20 min ultrasonication; Sample 3: MWCNTs−SDBS (1/1) nanofluid, wet grinding, 20 min ultrasonication; Sample 4: GN−SDS (1/1) nanofluid, wet grinding, 20 min ultrasonication; Sample 5: MWCNTs−SDS (1/1) nanofluid, wet grinding, 20 min ultrasonication; Sample 6: GN−SDS (2/1) nanofluid, wet grinding, 20 min ultrasonication)

It has been seen in Fig. 9 that both MWCNTs−SDS and MWCNTs−SDBS have increasing trends similar to Ref. [22], and later one has shown better thermal conductivity though both trends were bellow the thermal conductivity of distilled water in this experiment. However, for GN nanofluid, it has been seen that trend of GN−SDBS (sample 2) nanofluid went downward and is also below the trend of distilled water; whereas GN−SDS nanofluids (samples 4 and 6) showed retalively good thermal conductivity and 2/1 ratio solution gave better result than 1/1 ratio solution (sample 4) which supports the sedimentation test in this experiment, namely, lower amount of surfactant is better. As shown in Fig. 10, both sample 6 and sample 7 with different ultrasonication time (20 min and 40 min, respectively) gave thermal conductivity above that of distilled water. In addition, the increase in the ultrasonication time did not show better thermal conductivity. Though, sample 2 had positive percent enhancement at 25 °C as shown in Fig. 11, it was more negative at 35 °C. Sample 3 had a negative percent enhancement at both 25 °C and 35 °C. At 35 °C, it is almost four times. At 25 °C, sample 4, sample 6 and sample 7 had positive enhancements (4.202%, 5.546% and 4.706 %, respectively) and among them sample 6 gave better value. Similarly, at 35 °C, sample 6 also showed better value than other two.

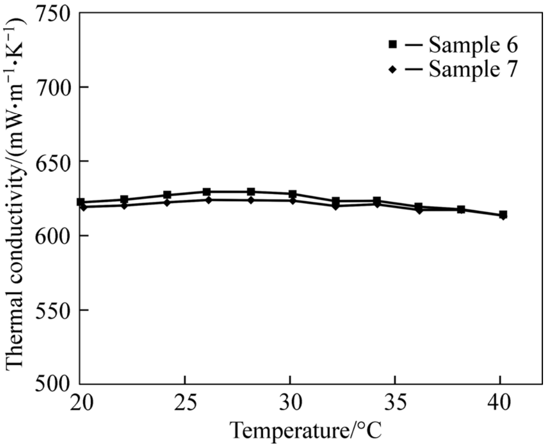

Fig. 10 Thermal conductivity vs temperature curves (Sample 6: GN−SDS (2/1) nanofluid, wet grinding, 20 min ultrasonication; Sample 7: GN−SDS (2/1) nanofluid, wet grinding, 40 min ultrasonication)

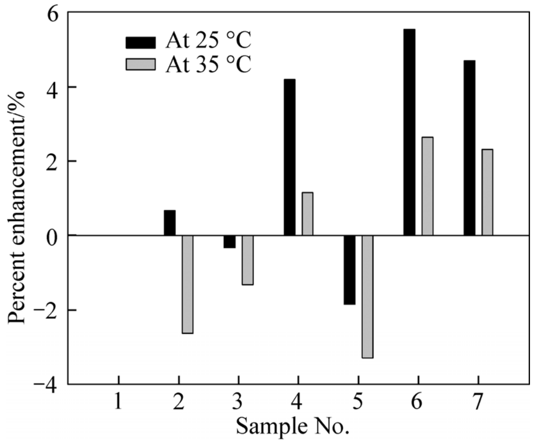

Fig. 11 Percent enhancement of thermal conductivities of nanofluids

From thermal conductivity analysis, it is obvious that the thermal conductivity was affected by four ways. First of all, amount of surfactant affected the thermal conductivity because absorbed surfactant molecules on the surface of the nanoparticle may increase the thermal resistance between nanoparticles and base fluid [37]. Secondly, chemical structure of surfactant modified the dispersibility of nanoparticles as graphene requires more planar surfactant than MWCNTs [27]. Thirdly, while grinding, there was a possibility of decomposition of organic surfactants that might lead to contamination despite wet grinding condition [7, 38]. Moreover, fourthly, ultrasonication of nanofluid might affect the surfactant used in this nanofluid [39] because ultrasonication can cause mechanical or chemical disruption to the surfactant molecules present in the nanofluid [27, 40].

3.4 Thermal effusivity

In thermodynamics, the thermal effusivity (e) of the material is defined as the square root of the product of the material's thermal conductivity (k) and its volumetric heat capacity [41]. It is the measurement of a material's ability to exchange thermal energy with its surroundings, i.e. the rate at which a material can absorb and transport heat. With a high thermal effusivity, the energy flux will also be high when there are temperature differences. The mathematical expression of thermal effusivity is

$$e=(k\rho c_p)^{1/2} \tag{1}$$

where k is the thermal conductivity i, ρ is density; c_p is specific heat capacity.

As shown in Fig. 12, both sample 6 and sample 7 had much higher effusivity than distilled water. So, these nanofluids can absorb and transport heat faster than distilled water. However, the effusivity curves have shown this possible only for a particular range of temperature. As shown in Fig. 12, with an increase in temperature, thermal effusivity of distilled water increased, whereas there was a gradual drop of sample 6 and sample 7. So, it is evident that for a particular temperature range, these two samples can be used as better coolant than water. However, at higher temperature this will not be applicable.

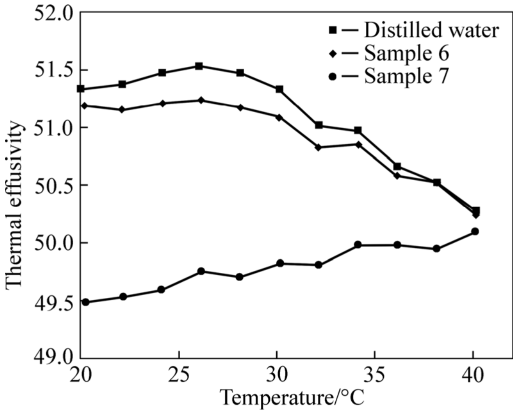

Fig. 12 Thermal effusivity

3.5 Thermal diffusivity

In heat transfer analysis, thermal diffusivity (α) is the thermal conductivity divided by density (ρ) and specific heat capacity (c_p) at constant pressure [42]. It is the measurement of how rapidly a material can adjust its temperature to that of its surroundings. It influences the penetration depth and the speed of the temperature adaption under a varying thermal state. In addition, it does not say anything about the energy flow. The SI unit is m^2/s, and the formula is:

$$\alpha=k/(\rho\cdot c_p) \tag{2}$$

where k is the thermal conductivity (W/(m·K)); ρ is density (kg/m^3); c_p is specific heat capacity (J/(kg·K)); together, ρc_p can be considered as the volumetric heat capacity ($J/(m^3\cdot K)$).

As shown in Fig. 13, sample 6 as the relalive higher thermal diffusivity which means that this nanofluid had a high conduction rate relative to its heat storage capacity and respond so faster than distilled water (DW) to change in temperature.

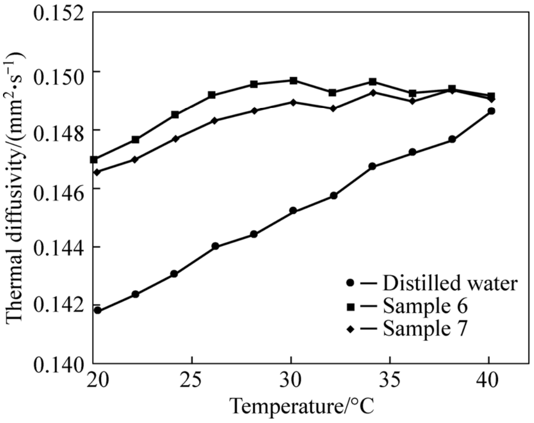

Fig. 13 Thermal diffusivity

3.6 Heat transfer coefficient enhancement ratio

Heat transfer coefficient is the calculation of heat transfer, typically by convection or phase transition between the fluid and a solid, and its SI unit is watts per square meter Kelvin ($W/(m^2\cdot K)$). Heat transfer coefficient depends on the fluid, the surface reference temperature and the flow regime. Considering convective heat transfer of laminar nanofluid flow inside a straight circular tube and constant wall temperature boundary condition, SADIK et al [43] explained that average heat transfer coefficient enhancement ratio equals one third power of volumetric heat capacity ratio of nanofluid to base fluid times two-thirds power of thermal conductivity ratio of nanofluid to base fluid, namely average heat transfer coefficient enhancement ratio increases by enhancing in volumetric heat capacity and thermal conductivity of nanofluid. In addition, there is a pronounced effect of thermal conductivity on heat transfer enhancement.

Heat transfer coefficient enhancement ratio is defined as the ratio of the heat transfer coefficient of the nanofluid to that of the base fluid [43]. As shown in Fig. 14, it has been easily seen that sample 6 had higher heat transfer coefficient enhancement ratio from 20 °C to 35 °C (about) than that of sample 7. However, with an increase in temperature, this value of these two samples decreased and after 35 °C these two values were almost same.

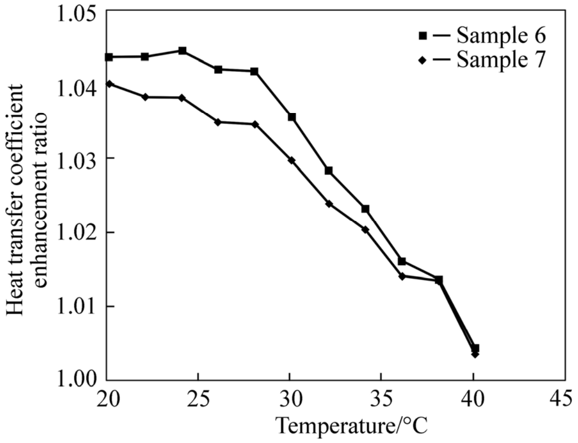

Fig. 14 Heat transfer coefficient enhancement ratio

4 Conclusions

1) GN nanofluids prepared with both surfactants (SDBS and SDS) with small adding ratios (2/1 and 3/1) by ultrasonication show long time stablity.

2) SDBS is helpful surfactant for dispersing MWCNTs, whereas SDS is effective surfactant for GN.

3) In this experiment, one of the causes of applying wet grinding is to minimize the possibility of contamination by decomposition of surfactants, and this is considered theoretically. The presence of surfactant induced contamination in grinding process can be disclosed by the analysis of X-ray diffraction (XRD) spectra where either change in peak positions or the appearance of new diffraction peaks will indicate the contamination. Shift in peak positions will show a change in lattice parameter and appearance of new diffraction peaks will indicate the formation of a new phase.

4) The decreasing trends of thermal conductivity are due to the sedimentation of nanoparticles in a base fluid (DW) with time.

5) Increasing the ultrasonication time for preparing GN-SDS nanofluid is not effective.

6) Besides the thermal conductivity, thermal diffusivity, thermal effusivity as well as heat transfer coefficient enhancement ratio are calculated and all of them revealed that GN−SDS with 2/1 ratio, 1 h with 500 r/min of wet grinding and 20 min ultrasonication is the better nanofluid by this solution approach.

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