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Thermal Conductivity of TiO_2 Nanoparticles Based Aqueous Nanofluids with an Addition of a Modified Silver Particle

Munkhbayar Batmunkh,[†,§] Md. R. Tanshen,[†] Md. J. Nine,[†,§] Munkhshur Myekhlai,[†] Heekyu Choi,[‡] Hanshik Chung,[†] and Hyomin Jeong[†,*]

[†]Department of Energy and Mechanical Engineering, Gyeongsang National University, Institute of Marine Industry, Cheondaegukchi-Gil 38, Tongyeong 650-160, Gyeongnam Korea

[‡]Engineering Research Center for Integrated Mechatronics Materials and Components, Changwon National University, Changwon 641-773, Korea

ABSTRACT: Nanofluid is a colloidal suspension which has received great attention over the past two decades, but its limited heat transfer enhancement is a matter of concern for industrial applications. We demonstrate an improvement in the thermal conductivity of TiO_2 nanofluids with an addition of negligible amounts of modified silver "Ag" nanoparticles. In this work, the surface/shape of newly synthesized "Ag" nanoparticles is modified by planetary ball milling. Then, to enhance the thermal conductivity of TiO_2 nanofluids, the flattened "Ag" particles are incorporated with the combination of small (15 nm) and large (300 nm) TiO_2 nanoparticles in an aqueous solution. The thermal conductivities of Ag/TiO_2−water nanofluids with various weight concentrations are measured at temperatures ranging from 15 to 40 °C. As a result, the present study confirms that the thermal conductivity of TiO_2 based solution can be improved by introducing the flattened "Ag" particles.

■ INTRODUCTION

When nanoscience and nanotechnology were speedily developing in 1995, nanometer (10^{-9} m) sized particles were dispersed in standard fluids; the resulting liquid was coined "nanofluid" by Choi.[1] Nanofluid is a new type of engineering fluid synthesized by suspending particles with a less than 100 nm size in common liquids such as water, ethylene glycol, and ethonal. The real applications of nanofluids are in photovoltaic devices and heating and cooling systems as well as in heat exchangers.[2−4]

To explore the thermal performance of fluids containing nanoparticles and use them in an applicatory place, it is important to deeply study their dispersion and thermal characteristics, which are critical thermo-physical properties. Therefore, researchers have investigated the dispersibility and thermal conductivity of various classes of nanoparticles, namely, (i) metallic particles,[5] (ii) nonmetallic particles,[6] and (iii) carbon allotropes[7,8] as additives in nanofluids. Among the nanofluids, TiO_2 nanoparticle based nanofluids have received much attention because of their wide range of applications, low cost, high thermal conductivity, and photocatalytic activity as well as dielectric properties.[9] The sample studies which showed the thermal behavior of TiO_2 nanoparticle based nanofluids are as follows.

Masuda et al.[10] analyzed the thermo-physical properties of the TiO_2 nanoparticles dispersed in an aqueous solution. A well known method (transient hot-wire) was applied to examine the thermal conductivity of samples. The authors found that the thermal conductivity of fluid containing nanoparticles was considerably higher than that of the base fluid. For instance, they revealed that around 11% higher thermal enhancement can be obtained for TiO_2 based aqueous nanofluids at 4.3 vol %, compared to the water-only.

Murshed et al.[11] studied the thermal characteristic of rod-shaped TiO_2 nanoparticles (10 nm in diameter × 40 nm in length) and spherical shaped TiO_2 (15 nm) that were suspended in water. As a result, a significant increment in thermal conductivity was observed with increasing the concentration of particles. In addition, it was found by these authors that the shape and size of the nanoparticles have an effect on the thermal characteristics of nanofluids.

He et al.[12] investigated the heat transfer characteristic of fluid containing TiO_2 nanoparticles in vertical tubes. The authors studied the viscosity and thermal conductivity of TiO_2 nanoparticle based aqueous nanofluids. The particle sizes of TiO_2 were 95, 145, and 210 nm with various concentrations (1−5 wt %). As a result, it was observed that small sized TiO_2 particles exhibited greater thermal conductivity than that of the large particles.

Until now, considerable attention has been paid to the synthesis of nanocomposites by incorporating different nanoparticles owing to their exceptional properties.[13−15] Recently, "Ag" nanoparticles have been mostly used as a supporting material to synthesize the composite materials[8,16] benefiting from its very high thermal and electrical conductivity.[5] "Ag" nanoparticles are also utilized in a wide range of practical applications such as in biosensors, biomedical imaging, Raman scattering, catalysts, as an antibacterial proxy, and in lithography.[17−22] However, no particular reports are available on the thermal characteristics of Ag/TiO_2 nanocomposite based nanofluids.

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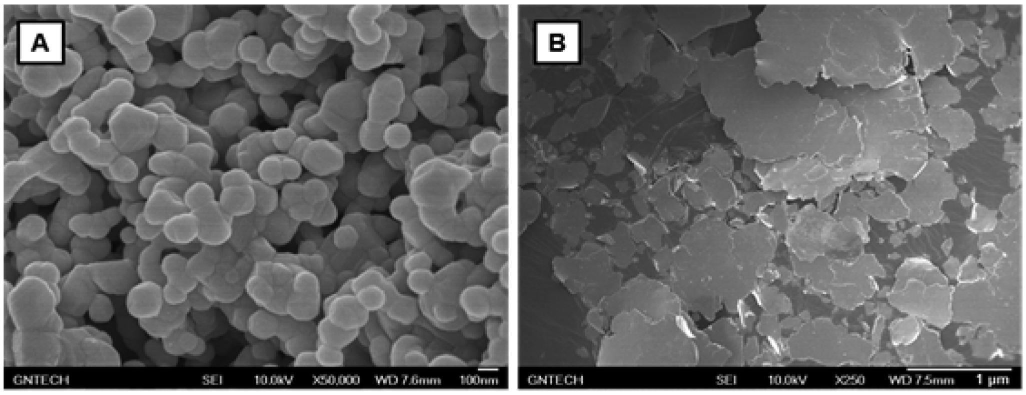

Figure 1. SEM images of (a) nonground and (b) ground "Ag" particles.

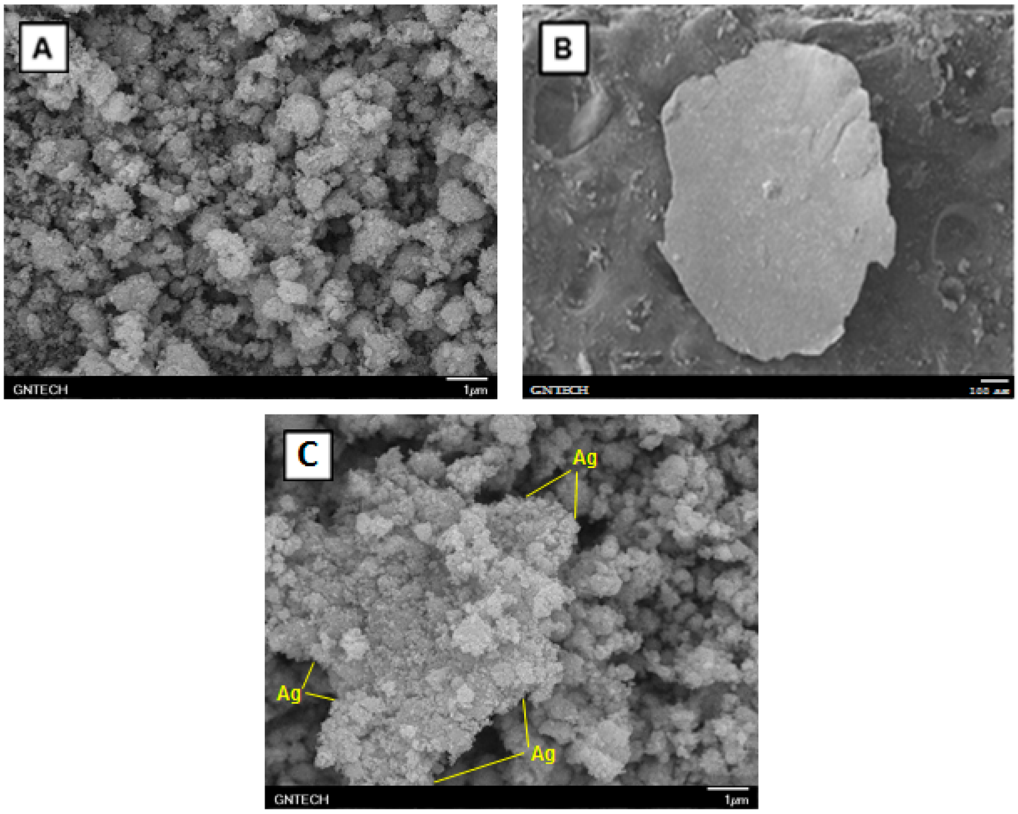

Figure 2. SEM images of the (a) combination of 70% small/30% large TiO_2, (b) the flattened "Ag" particle, and (c) Ag/TiO_2 composite.

Nonspherical structure based nanofluids are a proven concept for revealing greater thermal conductivity compared to the fluid containing spherical nanoparticles due to their higher aspect ratio.[5,23,24] In this study, newly synthesized "Ag" nanoparticles with spherical shape were ground using a planetary ball mill to change the particle shape and to increase the aspect ratio. Furthermore, to improve the thermal conductivity of TiO_2 nanofluids, we synthesized the Ag/TiO_2 nanocomposite based nanofluids by incorporating negligible amounts of flattened "Ag" structures into a combination of 70% small/30% large TiO_2 nanoparticles in water. It was believed that the flattened "Ag" particles and different sized TiO_2 particles could be effectively combined using ultrasonication.

■ EXPERIMENTAL DETAILS

Materials. "Ag" wire with a diameter of 0.2 mm and with a purity of 99.9% (obtained from Nano Technology Inc., Korea) was used to synthesize the Ag nanoparticles. Two different TiO_2 nanoparticles, namely ~15 nm TiO_2 with 99% purity (purchased from Sigma-Aldrich, MO, USA) referred to as small TiO_2 and ~300 nm TiO_2 with >97% purity (obtained from Sigma-Aldrich, MO, USA) referred as large TiO_2, were used to prepare the TiO_2 composite. As the base liquid, distilled water (DI water) was used to prepare the nanofluids.

Preparation of Nanofluids. First, the TiO_2 composite was prepared with a combination of 70% small (15 nm) and 30% large (300 nm) TiO_2 nanoparticles using a laboratory mechanical stirrer. For the comparison, three different aqueous nanofluids with 1, 2, and 3 wt % loadings of TiO_2 nanocomposite (combination of 15 and 300 nm sizes) were prepared by using a two-step method. For better dispersion, the powders were dispersed into the base fluids using an ultrasonication (Branson Corporation 41, USA) for 1 h. In the meantime, "Ag" nanoparticles were synthesized by a single-step approach using a pulsed-wire evaporation (PWE) method (received from Nano Colloid Maker, Nano Technology Inc., Korea). The working principle of this apparatus was clearly expressed in a previous study.[5] In this experimental work, we used a planetary ball mill (HPM-700) which was provided by Haji Engineering, Korea. Based on the previous work,[5] "Ag" nanoparticles were ground using a 1 mm ball size under wet conditions. In order to change the particle shape to be flattened, a rotation speed and a milling period were adjusted to be 500 rpm and 1 h, respectively. The pot rotation and the disk revolution were set as a counter direction. A detailed explanation of the ball milling process can be found elsewhere.[8] After grinding "Ag" nanoparticles, the flattened "Ag" nanoparticles are mixed with previously prepared TiO_2 composite. Then, Ag/TiO_2 composite particles with the flattened "Ag" structures were ultrasonically dispersed into the water. Ag/TiO_2 particle based nanofluids were prepared with three different concentrations, namely, (i) 1 wt % TiO_2 and 0.5 wt % "Ag, (ii) 2 wt % TiO_2 and 0.5 wt % Ag, and (iii) 3 wt % TiO_2 and 0.5 wt % Ag. Finally, we measured the thermal conductivities of six different nanofluids.

Instruments and Characterizations. The morphological structures and shapes of the particles were characterized by scanning electron microscopy (SEM; JSM-6710F, JEOL). The changes in structure and shape of particles were studied by SEM. The colloidal dispersibilities of the TiO_2 and Ag/TiO_2 particle based nanofluids were analyzed by a UV−vis spectrometer (X-ma 3000 Series Spectrometer, Human Co. Ltd., Korea) at wavelengths of 300−800 nm. X-ray diffractions (XRD) were carried out with a Bruker AXS, D8 advance powder diffractometer (Cu Kα radiation, λ = 1.5406 Å) in the 2θ range from 20° to 80°. The thermal conductivities of the prepared fluids were measured by a thermal conductivity analyzer (LAMBDA, Willingshausen, Germany) operating at temperatures ranging from 15 to 40 °C with an interval of 5 °C. The LAMBDA system analyzes the thermal characteristics of liquid samples according to ASTMD 2717. The operational principle of this technique is based on the transient hot wire method.[25,26]

■ RESULTS AND DISCUSSION

Materials Characterizations. Compared to a lower aspect ratio of nanoparticles, particles with a higher aspect ratio (L/D) exhibiting greater thermal and electrical conductivity is a reasonable concept.[27] Therefore, it was found in the literature[5] that the dispersion and thermal characteristics of metallic particles can be improved using a ball milling method. The ball milling can significantly change the shape of particles from spherical to flat. Therefore, this change in particle shape may affect the aspect ratio. Based on this respect, the synthesized "Ag" nanoparticles were ground using balls 1 mm in size. Figure 1 depicts the SEM images of newly prepared "Ag" nanostructures by the PWE method and ground "Ag" particles. SEM image of the synthesized "Ag" nanoparticles by the single-step method is shown in Figure 1a; the figure reveals that the particles are uniformly distributed and their shape is nearly spherical. The size of the synthesized "Ag" nanoparticles was about 100−120 nm. In addition, it can be observed that the synthesized particles possess a very smooth surface. Further-

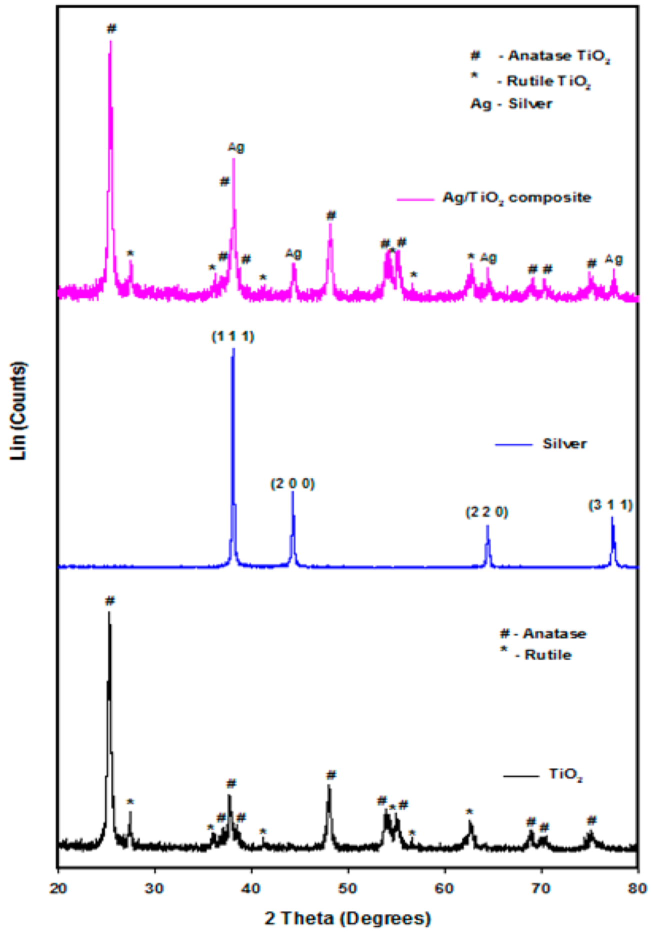

Figure 3. XRD patterns of the TiO_2, "Ag," and Ag/TiO_2 composite structures.

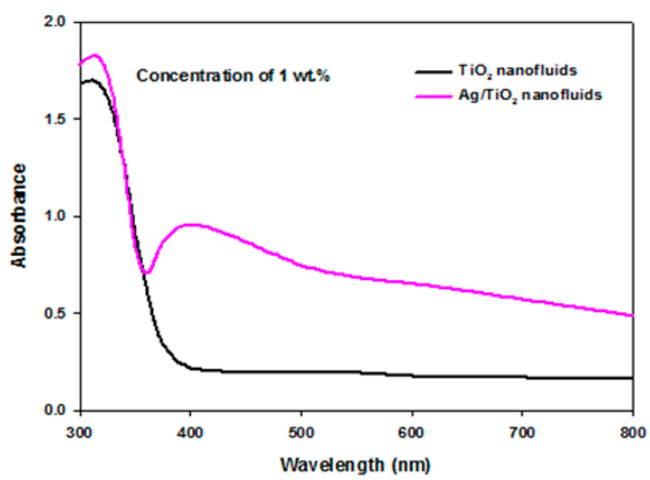

Figure 4. UV-absorption spectra of TiO_2 and Ag/TiO_2 structures with concentrations of 1 wt % based aqueous nanofluids.

more, the aspect ratio of these newly synthesized "Ag" nanoparticles can be estimated to be very low. This low aspect ratio of spherical nanoparticles would exhibit poorer thermal conductivity, compared to the particle with a high aspect ratio. As shown in Figure 1b, one can simply observe that the shape (spherical) of the synthesized "Ag" nanoparticles was completely changed to different sizes of flattened structures, after the application of ball milling. Such changes in particle

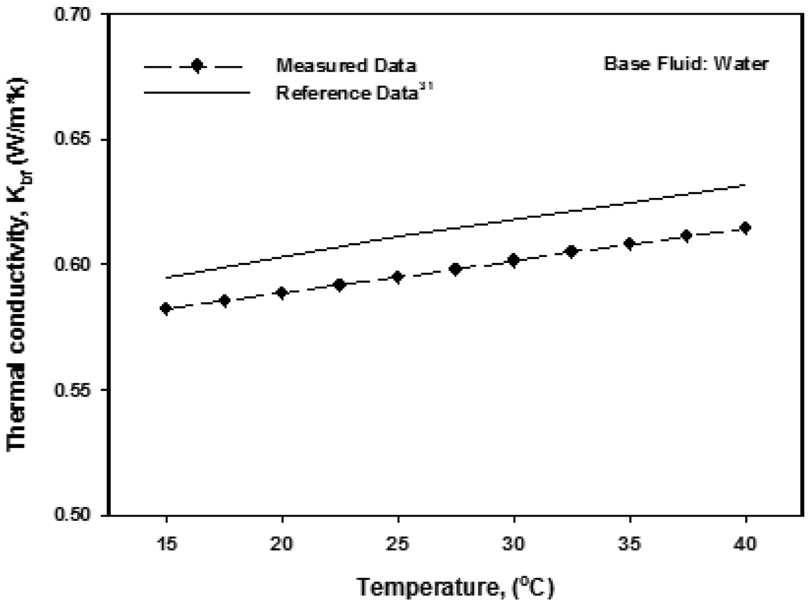

Figure 5. Measured thermal conductivity values of the base fluid (water) of this study and reference book.[31]

shape are probably due to the fact that small (1 mm) sized balls were strongly rolled out on the spherical particles. Based on these SEM images, the aspect ratio of the flattened particles can be estimated to be higher than that of the spherical particles.

The aim of the current study was to enhance the thermal conductivity of TiO_2 nanoparticle based aqueous nanofluids with an addition of a negligible amount of the flattened "Ag" particles. However, synthesis of the Ag/TiO_2 nanocomposite is the most essential to produce efficient nanofluids. To disperse the "Ag" particles into the TiO_2 nanoparticles, we prepared the TiO_2 composite with a combination of 70% small (15 nm) and 30% large (300 nm) TiO_2 nanoparticles. Figure 2a depicts the SEM image of the combination of 70% small and 30% large TiO_2 nanoparticles. It is seen from Figure 2a that the small TiO_2 particles with less than 25 nm are mixed with large TiO_2 particles. The majority of small TiO_2 particles also covered the large particles. Therefore, the formation of hollow cavities by these large TiO_2 particles and higher pore sizes can be clearly observed from Figure 2a. On the other hand, Figure 2b shows a high magnification of the SEM image of the flattened "Ag" particle. After the combination of these TiO_2 particles and "Ag" particles, several large TiO_2 particles, which were fully covered by the 15 nm TiO_2, were clearly decorated on the large sheet of "Ag" particles (see Figure 2c). It can be seen that the size and area of this "Ag" sheet is large. But, due to the presence of large TiO_2, it may be hard to find the small size of the flattened "Ag" particles in this Ag/TiO_2 composite structure.

To confirm the composition of the TiO_2 and the "Ag" particles, XRD analysis was conducted. Figure 3 clearly depicts the XRD patterns of the TiO_2, "Ag," and Ag/TiO_2 composite structures. As shown in Figure 3, several broad diffractions were assigned to the TiO_2 nanoparticles. Both anatase and rutile phases of TiO_2 bicrystalline structure were observed from the XRD result. Therefore, four narrow diffraction (1 1 1), (2 0 0), (2 2 0), and (3 1 1) peaks at $2\theta = 38.1°$, $2\theta = 44.2°$, $2\theta = 64.3°$, and $2\theta = 77.3°$, respectively, were observed for "Ag" nanostructures. Indeed, the XRD pattern of the Ag/TiO_2 composite structure showed all of the necessary characteristic of diffraction peaks of both TiO_2 and "Ag" at approximately $2\theta = 25.3°$, $2\theta = 27.4°$, $2\theta = 36.2°$, $2\theta = 38.06°$, $2\theta = 38.12°$, $2\theta = 38.4°$, $2\theta = 40.9°$, $2\theta = 44.2°$, $2\theta = 48.1°$, $2\theta = 53.9°$, $2\theta = 53.94°$, $2\theta = 54.9°$, $2\theta = 56.5°$, $2\theta = 62.7°$, $2\theta = 36.2°$, $2\theta = 64.3°$, $2\theta = 68.6°$, $2\theta = 70.6°$, $2\theta = 74.9°$, and $2\theta = 77.3°$. The XRD result suggests that the Ag/TiO_2 composite was successfully synthesized.

The effect of dispersion methods on the dispersibility of TiO_2 nanoparticle based aqueous nanofluids can also be characterized using a UV–vis spectrometer. It is possible to establish the existence of particles in the liquid phase based on the intensity of the corresponding absorption spectra. In addition, UV–vis absorption can be used to analyze the dynamics of the dispersion.[8,25] Higher absorbency means that the nanoparticles are well suspended and distributed in the base solution, indicating better dispersion stability of particles in the fluid.[28] UV–vis absorption spectra were recorded to measure the light absorption and the dispersion characteristics of suspensions. Figure 4 depicts the UV–vis spectra of TiO_2 and Ag/TiO_2 structure (1 wt %) based aqueous solution measured at wavelengths between 300 and 800 nm. It can be seen from Figure 4 that pure TiO_2 based solution has a broad intense absorption peak in the UV region (below 400 nm). This peak is a typical absorption of TiO_2 required to start electron transfer from the valence band (VB) to the conduction band (CB) of TiO_2. Interestingly, one can see that the result of the Ag/TiO_2 composite structure also shows intense and broad absorption at around 400–450 nm, which is a known characteristic of the surface Plasmon band for "Ag."[29] In addition, the result of UV–vis absorption also proves that the "Ag" particles were highly dispersed into the TiO_2 nanoparticles. Therefore, it has been demonstrated that the thermal characteristics of fluid containing nanoparticles strongly depend upon the dispersibility of nanofluids.[5,8,25,30]

Thermal Conductivity Analysis. Preparing a stable nanofluid is an important process to achieve a high thermal conductivity result. Therefore, the high thermal conductivity of fluids is one of most critical properties in their practical application. In this study, before analyzing the thermal characteristics of the prepared nanofluids, the instrument (Lambda system) was experimented with the base liquid (water). The measured value of the water was then compared

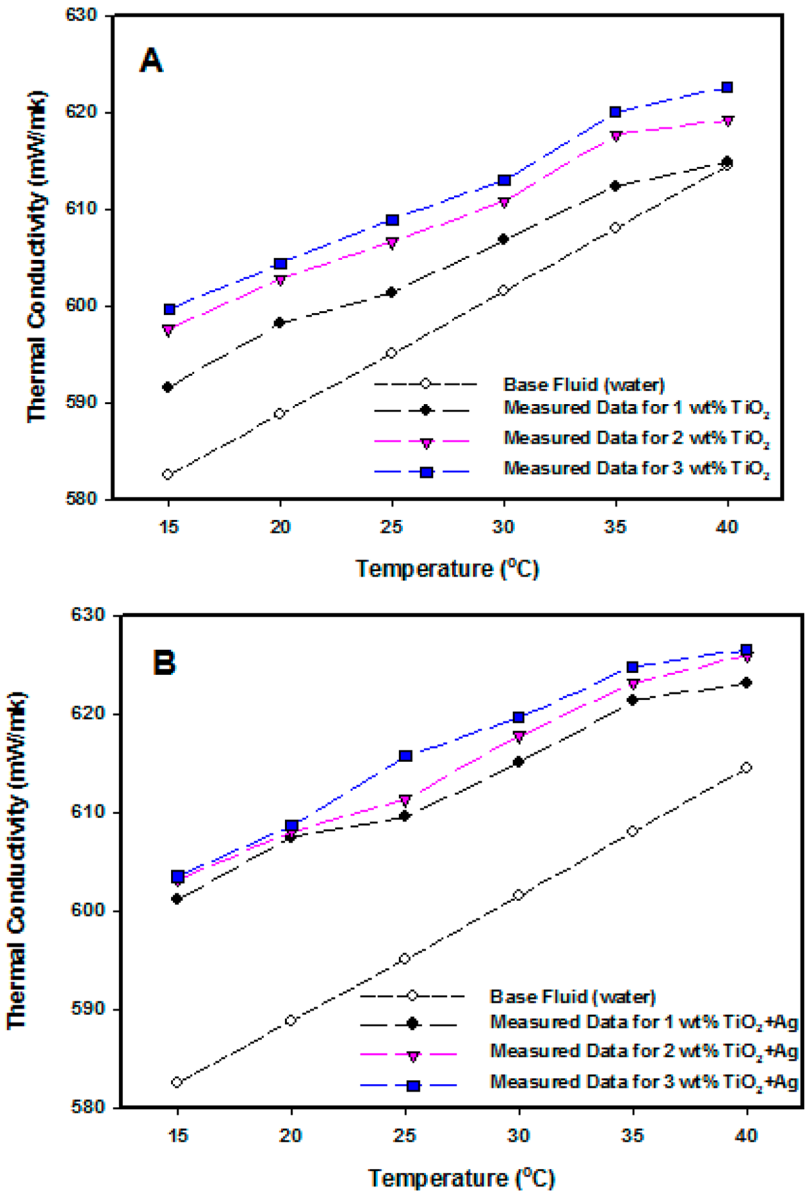

Figure 6. Thermal conductivities vs temperature of (A) TiO_2 and (B) Ag/TiO_2 nanofluids with varying loadings of TiO_2 nanoparticles. The results are compared to that of the water.

with the reference result displayed in a standard textbook.[31] Very good agreement between our data and reference data can be observed in Figure 5. The uncertainty in the measurement was only ∼2.0%.

It was expected that the combination of TiO_2 and "Ag" particles will enhance the thermal conductivity of fluids because "Ag" possesses high thermal conductivity. The temperature dependence of the thermal conductivities of TiO_2 and Ag/TiO_2 nanofluids with various concentrations of TiO_2 particles is illustrated in Figure 6. Therefore, Figure 6a and b show the thermal conductivities at a temperature ranging from 15 to 40 °C of only TiO_2 nanofluids and Ag/TiO_2 composite nanofluids, respectively. For better comparison, the measured values of the base fluid (water) were also plotted. It is seen from both Figure 6a and b that the thermal conductivities of the samples were linearly enhanced with raising the temperature. This phenomenon has been known to be due to the effect of enhanced Brownian motion. As the temperature rises, the viscosity of fluids apparently drops down, which improves the Brownian motion of the particles in base liquid, as explained by Jang and Choi.[32] It has been theoretically established that convection-like influences are developed by Brownian motion, which brings an enhanced thermal conductivity result. Another noticeable feature in Figure 6 is that conductivity of fluids containing nanoparticles improves with increasing the particle concentration in base fluid. This increment is due to the higher amount of particles suspended in the base liquid.[8,33]

To better realize the improvement in thermal conductivity of TiO_2 nanoparticle based fluids with the addition of "Ag" nanostructures, the thermal conductivities of TiO_2 and Ag/TiO_2 nanofluids were compared at a temperature of 25 °C (room temperature). Figure 7 depicts the thermal conductivities at a 25 °C temperature of the fluid containing only TiO_2 and Ag/TiO_2 composite as a function of TiO_2 loadings. To compare an increment in the thermal conductivities, the thermal conductivity data of the base liquid (water) was also plotted in Figure 7. The thermal conductivity of the water at 25 °C was measured to be 595.0 mW/m·K. As shown in Figure 7,

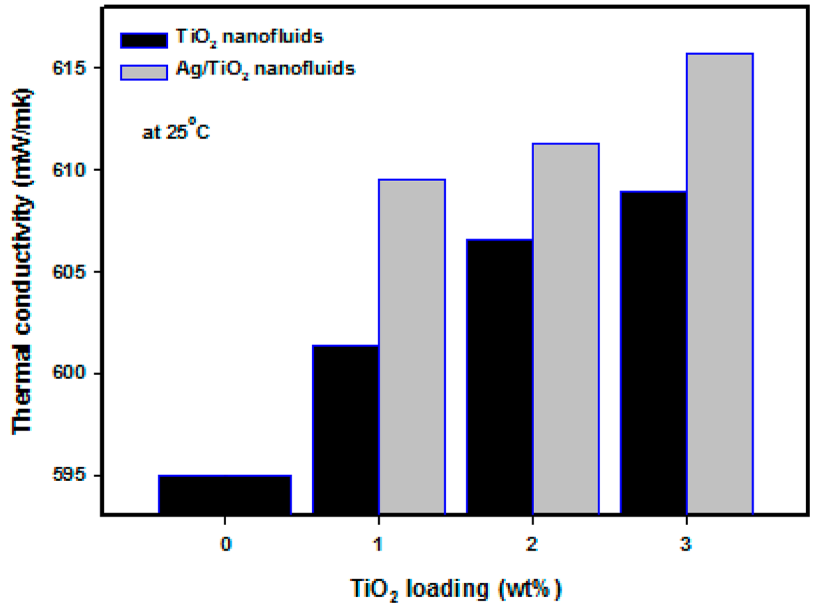

Figure 7. Thermal conductivity values of the fluid containing only TiO_2 and Ag/TiO_2 composite as a function of TiO_2 loadings. The results are compared to that of the water.

Table 1. List of Thermal Conductivities of Nanofluids (mW/m·K)

samples temperature (°C)	base fluid (water)	TiO_2 1 wt %	TiO_2 2 wt %	TiO_2 3 wt %	Ag/TiO_2 1 wt %	Ag/TiO_2 2 wt %	Ag/TiO_2 3 wt %
15	582.5	591.6	597.6	599.6	601.1	603.2	603.5
20	588.8	598.2	602.8	604.4	607.5	608.0	608.7
25	595.0	601.4	606.6	608.9	609.5	611.3	615.7
30	601.5	606.8	610.8	613.1	615.1	617.8	619.6
35	608.0	612.4	617.7	620.0	621.4	623.1	624.8
40	614.5	614.9	619.2	622.6	623.1	625.9	626.5

it can be apparently observed that Ag/TiO_2 composite structure based nanofluids exhibit a higher thermal conductivity than that of the TiO_2-only nanofluids. The thermal conductivity of TiO_2 nanocomposites based nanofluids with a concentration of 1 wt % is enhanced from 601.4 mW/m·K to 609.5 mW/m·K at room temperature (25 °C) after the addition of only 0.5 wt % Ag particles. This higher conductivity result was associated with the good dispersion of TiO_2 in the base fluid and the combination of TiO_2 and "Ag" structures. The combination of the particles improved the contact conductance of the TiO_2 nanoparticles. Because the "Ag" particle itself is a very good conductor (K_{Ag} = 429 mW/m·K), the interface of the Ag/TiO_2 composite positively facilitates phonon conduction by decreasing the boundary scattering failure[34] as well as interfacial resistance[35] between the particles and the fluids. Finally, Table 1 illustrates the thermal conductivity differences among the base fluid, TiO_2 nanofluids, and Ag/TiO_2 nanofluids.

CONCLUSION

The present paper describes an improvement in the thermal conductivity of TiO_2-water nanofluids by the addition of the flattened Ag structures. The surface/shape of the synthesized Ag nanoparticles was changed to be flattened by planetary ball milling. From the results, X-ray diffraction (XRD) and UV−vis spectrometer results confirm that the Ag/TiO_2 composite was successfully synthesized. The thermal conductivity of TiO_2-water nanofluids with a concentration of 1 wt % was enhanced from 601.4 mW/m·K to 609.5 mW/m·K at room temperature (25 °C) after the addition of only 0.5 wt % Ag particles. This enhancement was associated with the interface of Ag/TiO_2 composite positively facilitating phonon conduction by decreasing the boundary scattering failure[34] as well as interfacial resistance[35] between the particles and the fluids.

AUTHOR INFORMATION

Corresponding Author

*Tel.: +82 055 640 3184. E-mail: hmjeong@gnu.ac.kr.

Present Address

§School of Chemical Engineering, The University of Adelaide, Adelaide, South Australia 5005, Australia

Notes

The authors declare no competing financial interest.

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