

# Effect of Ultrasonication Duration on Colloidal Structure and Viscosity of Alumina–Water Nanofluid

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**ABSTRACT:** Nanofluids are promising fluids for heat-transfer applications. Low stability and high viscosity are two important drawbacks for practical applications of nanofluids. The aggregation and sedimentation of nanoparticles are related to the colloidal structure of nanofluids, which directly affects the stability and viscosity. An ultrasonic homogenizer can break the aggregation of particles. The aim of this work was to study the effect of the duration of ultrasonic treatment on colloidal structure, including the stability and temperature-dependent viscosity of a nanofluid. Specifically, a 0.5 vol %  $\text{Al}_2\text{O}_3$ –water nanofluid was prepared using an ultrasonic homogenizer for various durations from 0 to 180 min. The microstructure, colloid and particle sizes, precipitation, and zeta ( $\zeta$ ) potential were analyzed to investigate the aggregation and sedimentation of the nanofluid. The viscosities of nanofluids subjected to ultrasonic treatment for different durations were also measured at different temperatures from 15 to 45 °C. Better particle dispersion, lower particle sizes, smaller colloid sizes, less precipitation, and higher  $\zeta$  potentials were observed with increasing sonication time. The viscosity of  $\text{Al}_2\text{O}_3$ –water nanofluid was found to increase with the sonication time up to 60 min and then subsequently decreased. In addition, the viscosity decreased with increasing temperature. The research concluded that more stabler and lower-viscosity nanofluids can be obtained by applying ultrasonic treatment for durations of 90 min or longer.

## 1. INTRODUCTION

Heat-transfer enhancement is emphasized for energy-saving purposes that could lead to the better quality of human life and meet the aim of sustainable development. Nanotechnology plays a vital role in heat-transfer enhancement. Fluids that contain nanoparticles are called “nanofluids”.<sup>1</sup> Nanofluids are colloidal suspensions of nanoparticles (with an average particle size of less than 100 nm in at least one dimension) dispersed in base fluids such as water or glycols to enhance their thermal performance.<sup>2,3</sup> The stability of nanofluids is a critical factor that must be taken into account because it affects the performance of any system. Nanoparticles tend to agglomerate easily over time because of their high surface energies. The aggregation of nanoparticles is a reason for sedimentation, which decreases the thermal conductivity of nanofluids.<sup>4</sup> In addition, the sizes of nanoparticle agglomerates also affect the viscosity of nanofluids and, consequently, lead to lower heat-transfer performance.<sup>5</sup>

Ruan and Jacobi<sup>5</sup> reported that ultrasonication is a common way to break up agglomeration and promote the dispersion of nanoparticles into base fluids to obtain more stable nanofluid. Ultrasonication techniques affect the surface and structure of nanoparticles and prevent the agglomeration of particles to achieve stable nanofluids.<sup>6</sup> Addition of surfactant is another method that is used to increase the stability of nanofluids. Surfactants, also known as surface-active agents, are chemical compounds that reduce the surface tension of a liquid and increase the immersion of particles. Use of a surfactant is necessary for insoluble particles such as carbon nanotubes (CNTs) that do not disperse in most solvents.<sup>7</sup> However, some surfactants, such as gum arabic (GA), increase the viscosity of

nanofluids, causing an increase in pressure drop and pumping power, especially in industrial applications.<sup>8</sup>

Thus, ultrasonication methods are popular among researchers. Nevertheless, there is no standard procedure for the ultrasonication process to prepare a nanofluid as very little research has been done on the effect of the duration of ultrasonic treatment. Therefore, researchers are struggling to prepare stable and well-dispersed nanofluids as a result of viscosity increases and discouraging researchers to use these nanofluids in any system. Moreover, inconsistent outcomes have been reported in the literature even for the same type of nanofluid because of the lack of the standard preparation process. Very few comparative studies on the optimum sonication time required to prepare stable nanofluids are available in the literature. Kwak and Kim<sup>9</sup> studied the optimum ultrasonication duration for CuO–ethylene glycol (EG) nanofluid. They ultrasonicated the mixture for between 1 and 30 h and characterized the resulting nanofluid by dynamic light scattering (DLS) and zeta ( $\zeta$ ) potential measurements. They found that 9 h of sonication was optimum and that, after longer sonication, the particles coalesced again. Lee et al.<sup>10</sup> ultrasonicated  $\text{Al}_2\text{O}_3$  nanoparticles in water for durations of 0, 5, 20, and 30 h. Transmission electron microscopy (TEM) and  $\zeta$  potential measurements were used to characterize the nanofluid. It was found that a sonication duration of ~5 h gave the best results. Garg et al.<sup>8</sup> ultrasonicated a suspension of CNTs in deionized water for up to 80 min and found that the optimum

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ultrasonication time for homogenization was 40 min. However, Amrollahi et al.<sup>11</sup> applied ultrasonication for 20 h, and Ruan and Jacobi<sup>5</sup> applied up to 1355 min to homogenize CNTs in EG. Both studies reported that longer sonication times gave better results. Yu et al.<sup>12</sup> studied the dispersion behavior of CNTs under varying sonication times. They prepared nanofluids with 0.1 wt % multiwalled carbon nanotubes (MWCNTs) in distilled water with 0.15 wt % sodium dodecyl sulfate (SDS) as the surfactant. A continuous power of 20 W was used for sonication with various durations of 0, 5, 15, 30, 40, 50, and 120 min. They characterized the colloid with UV-visible spectrophotometer and TEM and found that more energy (sonication time) was required to disperse a higher amount of particles in solution. Chung et al.<sup>13</sup> dispersed two types (A and B) of ZnO nanopowder in water and ultrasonicated the dispersions for 60 min. They characterized the effect of various sonication times using TEM and photocorrelation spectroscopy (PCS). The PCS results showed that ultrasonication reduced the mean cluster size to 100 nm within 60 min for powder A and within 20 min for powder B, whereas further ultrasonication up to 60 min could not reduce the cluster size. Nevertheless, the TEM results showed that aggregates still existed in the suspension.

It would be better if the physical properties could be measured with the colloidal structure for the effect of ultrasonication duration to confirm the relationship between cluster size and physical properties. Amrollahi et al.<sup>11</sup> measured the thermal conductivity of CNTs in EG after different ultrasonication durations up to 24 h. They reported that higher sonication time gave higher thermal conductivity ratios and that this enhancement was more significant for higher concentrations. Ruan and Jacobi<sup>5</sup> investigated the effects of sonication on the thermal conductivity and viscosity of 0.5 wt % CNTs in EG. They indicated that the maximum enhancement in thermal conductivity and the minimum increase in viscosity were obtained using longer sonication times. They found that the maximum viscosity rise of the nanofluid was obtained after a sonication time of 40 min, finally decreasing to the viscosity of the pure base fluid level at a sonication time of 1355 min. Garg et al.<sup>8</sup> investigated the effects of sonication time on the thermal conductivity and viscosity of nanofluids. The authors prepared four samples of 1 wt % MWCNTs in GA and water and subjected the samples to ultrasonication for 20–80 min. They found that the thermal conductivity increased with the ultrasonication duration but the viscosity initially increased until 40 min and then decreased with further increasing sonication time. However, Yang et al.<sup>14</sup> found that the viscosity of nanofluids kept decreasing with increasing dispersion energy for nanotube-in-oil dispersions and that the sonication time was proportional to the dispersion energy.

From the above limited literature, no concrete conclusions can be drawn. Nevertheless, in the case of thermal conductivity, researchers agree fact that it increases with increasing sonication time. However, in both cases (colloidal structure and viscosity), some researchers have found that the highest sonication is best, whereas others have reported an optimum ultrasonication duration. Therefore, more studies are needed to specify the required ultrasonication time for preparing more stable and less viscous nanofluids. Furthermore, most studies are concerned with CNT nanofluids because of their high thermal conductivity despite some drawbacks such as being insoluble in most liquids and having high costs and a difficult manufacturing process. The effect of ultrasonication is

significant in CNT nanofluids because of the high aspect ratio (length-to-diameter ratio) of CNTs. The length usually tends to break down because of the force created during the sonication period. Al<sub>2</sub>O<sub>3</sub> is a potential nanoparticle as it disperses easily in most fluids. It is inexpensive, as its manufacturing is easy and it is produced on a large industrial scale. Because alumina has a spherical shape in most cases, it has a minimum aspect ratio (about 1). Moreover, to the best of our knowledge, no study on the effects of ultrasonication duration on the viscosity of Al<sub>2</sub>O<sub>3</sub>-based nanofluids is available. Therefore, it is necessary to analyze the effects of ultrasonication on colloid and the thermophysical properties of Al<sub>2</sub>O<sub>3</sub>-based nanofluids. Thus, in this study, the effects of ultrasonication duration on the colloidal structure and viscosity of 0.5 vol % Al<sub>2</sub>O<sub>3</sub>-water nanofluid have been studied.

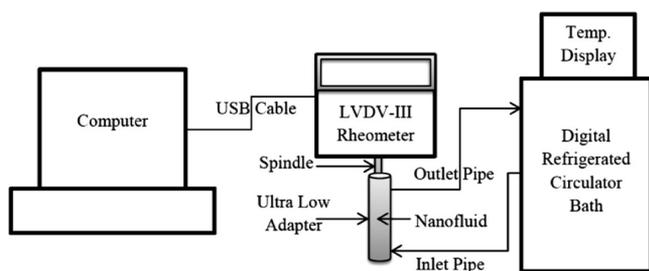
## 2. EXPERIMENTAL METHOD

**2.1. Nanofluid Preparation.** Al<sub>2</sub>O<sub>3</sub> nanoparticles (manufactured by Sigma-Aldrich, St. Louis, MO, and directly purchased from Sigma-Aldrich, Selangor Darul Ehsan, Malaysia) with an average particle size of 13 nm and a purity of 99.5% were used in this study. Distilled water was used as the base fluid for these experiments. A two-step method was employed to prepare this nanofluid, where nanoparticles were primarily arranged and then mixed with the fluid using ultrasound.<sup>15</sup> First, 0.5 vol % Al<sub>2</sub>O<sub>3</sub> nanoparticles were suspended in distilled water to prepare the nanofluid. After the nanoparticles had been suspended in the base fluid, the mixtures were stirred by a very thin and narrow metal strip for 1 min to enable the nanoparticles to subside into the base fluid completely. Then, the nanofluids were sonicated for durations of 30, 60, 90, 120, 150, and 180 min using an ultrasonic homogenizer machine (model 505, Fisher Scientific, Pittsburgh, PA). Pulses with a 50% amplitude and a sequence of 2 s on and 2 s off were used during the sonication process. Ultrasonication could affect the total volume and concentration of nanofluids as the agitation increase the temperature by 10 °C/min initially.<sup>13</sup> Therefore, an advanced digital refrigerated circulator bath (model AD07R-40-12E, PolyScience, Niles, IL) was connected to a recursion beaker, and nanofluids were prepared inside the beaker at 15 °C to avoid vaporization.

**2.2. Colloid Inspection.** The microstructure and composition of the nanoparticles were characterized by field-emission scanning electron microscopy (FESEM) (model AURIGA, Carl Zeiss, Oberkochen, Germany). As-received nanoparticles were characterized by FESEM at 1 kV kinetic energy without any treatment. Magnification scales of 1000× and 10000× were used to capture the image within 10- and 1-μm plots, respectively. TEM (model LIBRA 120, Carl Zeiss, Oberkochen, Germany) was used (with 120 kV kinetic energy) to capture the microstructures of the nanofluid for the purpose of analyzing the colloidal structure. Samples for TEM were prepared immediately after the preparation of the nanofluid in a transparent and thin copper grid. Magnification scales of 6300× and 31500× were used to capture the image within 500- and 50-nm plots, respectively. The average particle diameter for all ultrasonication durations was also measured as an arbitrary distance by TEM. At least two images on the 50-nm scale were analyzed for each sample, and the diameters of 10–12 nanoparticles were measured to check the particle size after each sonication. A Zetasizer 3000HS instrument (Malvern Instruments, Malvern, U.K.) was used to check the average colloid size and ζ potential after sonication of each sample. The

Zetasizer analysis was conducted at 25 °C 24 h after nanofluid preparation. The samples “passed” the quality factor during the first run even without diluting the concentration. The  $\zeta$  potential was analyzed without changing the pH of the suspension. Precipitation of the prepared nanofluids was investigated using the sedimentation photocapturing method. After preparation, 16-mL samples of nanofluids ultrasonicated for different durations were poured into small bottles. Photographs of the bottles containing the nanofluids were captured 30 days after preparation for stability inspection.

**2.3. Viscosity Measurement.** In this study, a programmable rheometer (model LVDV-III ultra, Brookfield, Middleboro, MA) was used to measure the viscosity. The machine was connected to a personal computer via USB cable, and Rheocalc 32 software was used for data collection and storage. The spindle was connected to the viscometer and submerged into the nanofluid. The viscosity developed against the spindle as a result of deflection of the calibrated spring with the range of viscosity between 1 and  $6 \times 10^6$  mPa·s with the aid of an ultra low adapter (ULA) attached to the main equipment.<sup>16</sup> The viscosity of each sample was measured at a constant shear rate of  $122 \text{ s}^{-1}$  while the ULA spindle was rotating at 100 rpm. For the temperature variation, the refrigerated circulator bath (model AD07R-40-12E, PolyScience, Niles, IL) was connected to the water jacket of the ULA that was attached to the rheometer. The temperature of each sample was varied from 15 to 45 °C at 10 °C intervals to investigate the effect of temperature on the viscosity of the nanofluid. The precision of temperature measurements was within the range of  $\pm 0.4$  °C. A schematic of the viscosity measurement system is shown in Figure 1. Each experiment was repeated three times to obtain



**Figure 1.** Schematic illustration of the viscosity measurement apparatus.<sup>17</sup>

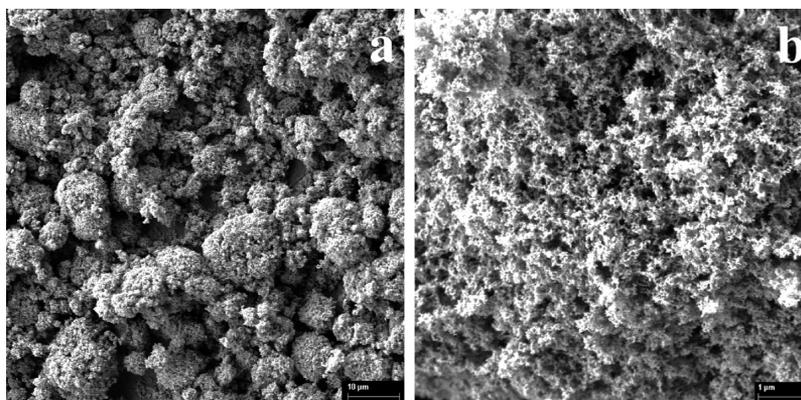
values that were more precise. The mean value of the three data points was considered for the analysis. The average uncertainty of viscosity measurements was found to be  $\pm 1.6\%$ .

### 3. RESULTS AND DISCUSSION

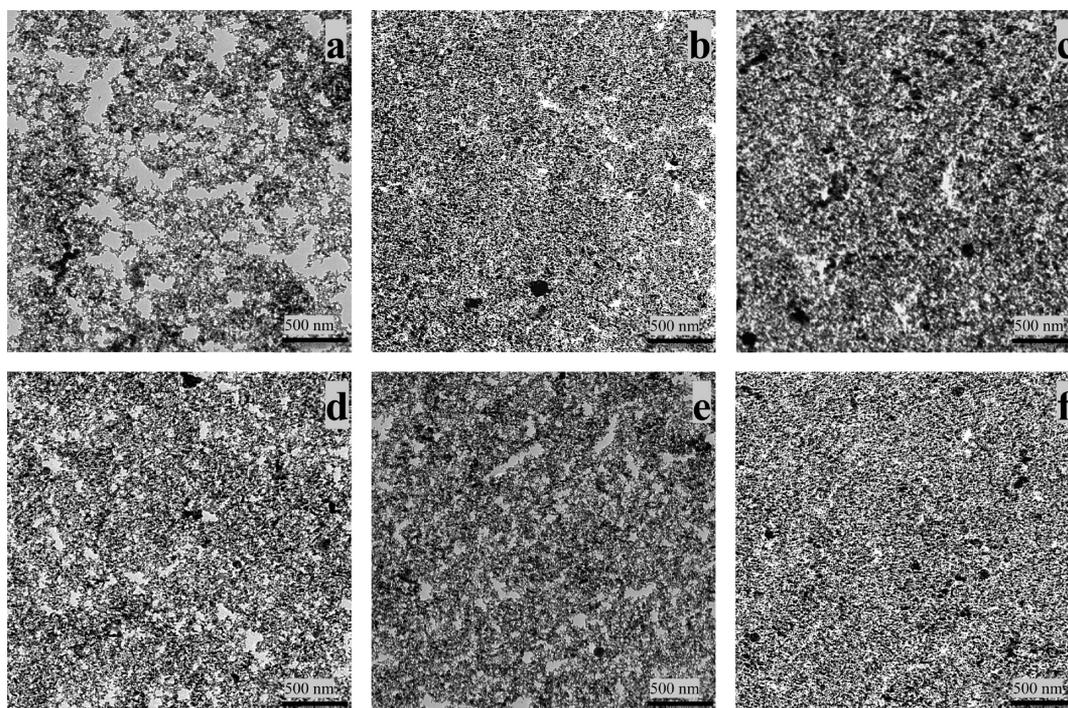
#### 3.1. Effect of Ultrasonication on Colloidal Structure.

To study the effect of sonication time on the colloidal structure of  $\text{Al}_2\text{O}_3$ –water nanofluid, the microstructure of  $\text{Al}_2\text{O}_3$  nanoparticles was observed first, before they were mixed with water. After the  $\text{Al}_2\text{O}_3$  nanoparticles had been suspended in water, the microstructure was again analyzed by TEM. The microstructures of  $\text{Al}_2\text{O}_3$  nanoparticles taken by FESEM without any treatment (as received) are shown in Figure 2. In Figure 2a, in the 10- $\mu\text{m}$  range, high agglomeration of nanoparticles is observed. Figure 2b shows the particles in the smaller range of 1  $\mu\text{m}$ , in which the nanoparticles are found in loose clustered form and spherical/cylindrical shape.

The TEM images of  $\text{Al}_2\text{O}_3$ –water nanofluid after each sonication are shown in Figure 3. To obtain a better understanding of the colloidal structure, a wide range (500-nm scale) was chosen here. Figure 3a shows the microstructure of  $\text{Al}_2\text{O}_3$ –water nanofluid after 30 min of ultrasonication. From Figure 3a, it is clear that the nanoparticles were not well dispersed and there was strong clustering among the nanoparticles. Some locations that are empty in the micrograph imply the presence of no particles, whereas some places are darker and show high aggregation of nanoparticles. Figure 3b (for 60 min of ultrasonication) shows the good dispersion of nanoparticles, with minor overlap of nanoparticles but some empty areas. Figure 3c (for 90 min of ultrasonication) is similar to Figure 3b, but more spreading of nanoparticles is seen with few empty spaces. However, some nanoparticles still overlap. Figure 3d (for 120 min of ultrasonication) shows that nanoparticles coalesced again, which is even more obvious in Figure 3e (for 150 min of ultrasonication). For further ultrasonication of 180 min, the micrograph shows a very good dispersion of nanoparticles (see Figure 3f). Therefore, a strong morphological change in the colloid occurs with the variation of the ultrasonication duration, as nanoparticles are dispersed until 90 min of sonication after which they start to coalesce and are again homogeneously dispersed with 180 min of ultrasonication. Some more related microstructures are shown in Figure 5 that provide more evidence in support of the above statement. Kwak and Kim<sup>9</sup> found a similar type of morphology, as further sonication after the optimum sonication time caused the nanoparticles to coalesce again.

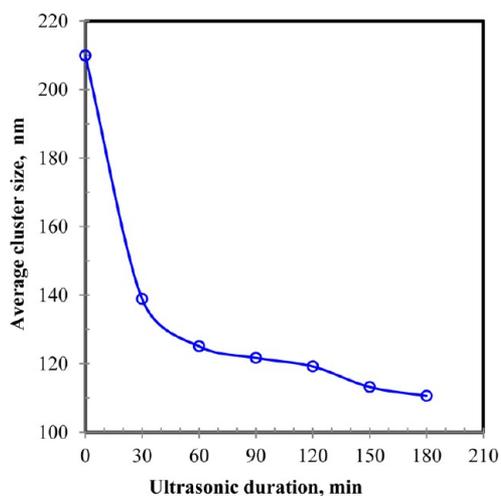


**Figure 2.** FESEM images of  $\text{Al}_2\text{O}_3$  nanoparticles at (a) 10- and (b) 1- $\mu\text{m}$  scales.



**Figure 3.** Microstructure of  $\text{Al}_2\text{O}_3$ –water nanofluid after (a) 30, (b) 60, (c) 90, (d) 120, (e) 150, and (f) 180 min of ultrasonication.

There is uncertainty in the microstructure of nanofluids taken by TEM because this technique analyzes a very small amount of sample. Therefore, a Zetasizer instrument was used to measure the average size of aggregation using the photon correlation spectroscopy (PCS) method. The effect of ultrasonication duration on the average cluster size is reported in Figure 4. It can be seen in Figure 4 that the aggregation size



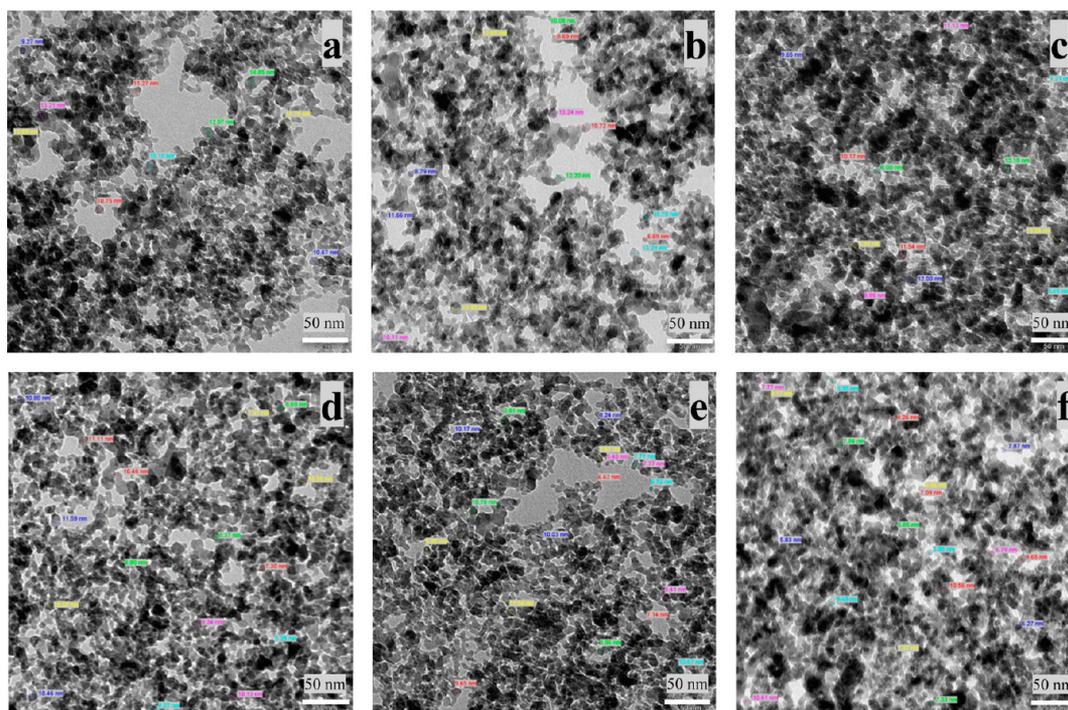
**Figure 4.** Average cluster size of  $\text{Al}_2\text{O}_3$  nanoparticles after different durations of ultrasonication.

decreased with increasing sonication time. The average cluster size rapidly decreased with the start of sonication. Initially, the decreasing rate of aggregation size was found to be higher. After a certain duration, the rate of decrease was lower. In this study, the average cluster size decreased from 210 nm (for 0 min, i.e., without ultrasonication) to 125 nm with 60 min of ultrasonication. However, with further ultrasonication, the average

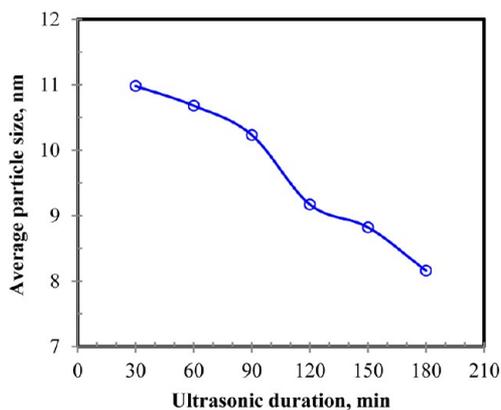
cluster size was found to be 113 and 110 nm for 150 and 180 min of sonication, respectively.

The final particle sizes (average) of  $\text{Al}_2\text{O}_3$ –water nanofluid after each ultrasonication were also measured from TEM images, as shown in Figure 5 (on a smaller scale as 50 nm). The morphological changes described in Figure 3 are again clear in Figure 5. Figure 5a shows the results after 30 min of ultrasonication, where the nanoparticles were not well dispersed and strong clustering among the nanoparticles is clearly observed. Figure 5b (for 60 min of ultrasonication) shows that nanoparticles were not well dispersed but less agglomeration and very few overlapping of nanoparticles occurred. Figure 5c (for 90 min of ultrasonication) shows a good dispersion of nanoparticles but still some agglomeration. According to Figure 5d (for 120 min of ultrasonication), the nanoparticles coalesced again. This is even more obvious in Figure 5e (for 150 min of ultrasonication). Again, for 180 min of ultrasonication, a more homogeneous dispersion of nanoparticles is shown in Figure 5f. The final particle size after each sonication is plotted in Figure 6. It can be seen in Figure 6 that the average nanoparticle size decreased with increasing ultrasonication duration. An almost linear decreasing trend of particle size was observed, and the average final particle size was found to be 8 nm after 180 min of ultrasonication. Therefore, this study supports the statement of Yang et al.,<sup>14</sup> who reported that prolonged ultrasonication time affects the size and aspect ratio of particles, which is more significant for nanotubes because of their larger particle length.

The stability of  $\text{Al}_2\text{O}_3$ –water nanofluid for different ultrasonication durations was checked by capturing photographs 30 days after preparation, as shown in Figure 7. A thick layer of sedimentation formed at the bottom of the nanofluid without ultrasonication (0 min), as shown in Figure 7a. However, for the nanofluids prepared by the ultrasonication process, the amount of sedimentation was found to be negligible, as can be seen in Figure 7b–g. Supernatant levels are marked in Figure 7,



**Figure 5.** TEM microstructure of  $\text{Al}_2\text{O}_3$ –water nanofluid after (a) 30, (b) 60, (c) 90, (d) 120, (e) 150, and (f) 180 min of ultrasonication on a 50-nm scale with particle size measurements.

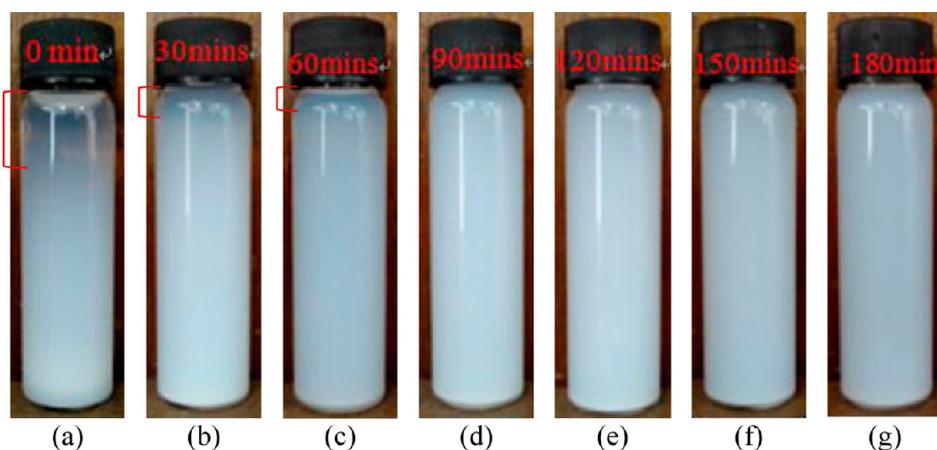


**Figure 6.** Average final particle size of  $\text{Al}_2\text{O}_3$  nanoparticles after different durations of ultrasonication.

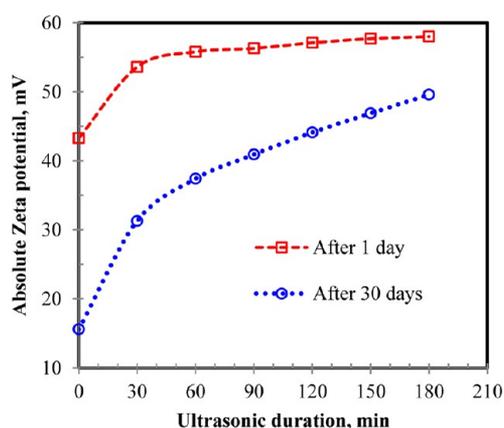
although it was very difficult to determine the exact level. The supernatant level was found to be higher in Figure 7a, which confirms the higher sedimentation of particles. Panels b and c of Figure 7 show a very low supernatant level, whereas in Figure 7d–g, no supernatant can be observed. This indicates that the stability of the nanofluid increased with increasing ultrasonication duration. Therefore, the stability of the nanofluids can be improved by increasing the ultrasonication duration during their preparation. This method (photographic analysis) is a qualitative analysis, and exact quantities could not be measured. Therefore, the  $\zeta$  potential was measured for each sample to quantify the stability of the nanofluid, as shown in Figure 8. It is pronounced that absolute  $\zeta$  potential values over 60 mV indicate excellent stability, those above 30 mV indicate physical stability, those below 20 mV indicate limited stability, and those lower than 5 mV are evidence of agglomeration.<sup>18</sup> In this study, the absolute  $\zeta$  potential value increased with ultrasonication duration as seen in Figure 8. With the starting of

ultrasonication,  $\zeta$  potential increases to a higher value and further ultrasonication it rises slowly. Highest  $\zeta$  potential value was found to be 58 mV for 180 min of ultrasonication. Kwak and Kim<sup>9</sup> found the highest absolute  $\zeta$  potential value of about 50 mV for 9 h of sonication, whereas Lee et al.<sup>10</sup> found a value of about 34.5 mV for 5 h of sonication. A comparison was drawn between the values of the  $\zeta$  potential after 1 day and after 30 days of preparation. It was observed that, after 30 days, the  $\zeta$  potential values decreased, and the difference between 1 day and 30 days was higher for lower sonication time. Thirty days after preparation, the absolute value was found to be 15 mV for the sample without sonication (termed as 0 min). That is why, in Figure 7a, the supernatant level was found to be higher for 0 min due to the lower  $\zeta$  potential value. Even after 30 days of preparation, the absolute  $\zeta$  potential value was found to be 49 mV for an ultrasonication duration of 180 min, and in Figure 7g, no sedimentation or supernatant level can be observed. Therefore, the absolute  $\zeta$  potential value is fully related to the stability of the nanofluid, and longer sonication durations increased the stability of the nanofluid.

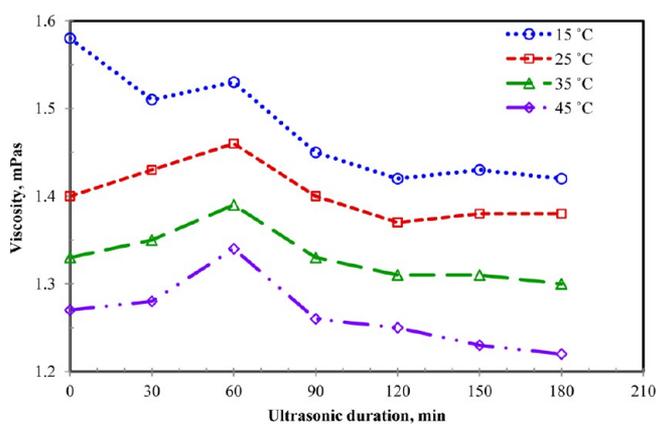
**3.2. Effect of Ultrasonication on Viscosity.** The effects of temperature and ultrasonication duration on the viscosity of 0.5 vol %  $\text{Al}_2\text{O}_3$ –water nanofluid are shown in Figure 9. It can be seen that the viscosity of the ultrasonicated nanofluid significantly decreased as the temperature was increased from 15 to 45 °C. The decrease in the viscosity with increasing temperature is due to the weakening of interparticle adhesion forces. When temperature increases, the heat energy provides extra energy to separate the molecules, resulting in the reduction of attractive forces between molecules. A higher nanofluid temperature intensifies the Brownian motion of the nanoparticles and reduces the viscosity of the nanofluid.<sup>19</sup> At 15 °C, the viscosity of the  $\text{Al}_2\text{O}_3$ –water nanofluid without ultrasonication was the highest compared to those of the other ultrasonicated nanofluids. This can be explained by the



**Figure 7.**  $\text{Al}_2\text{O}_3$ -water nanofluids prepared with ultrasonication durations of (a) 0, (b) 30, (c) 60, (d) 90, (e) 120, (f) 150, and (g) 180 min 30 days after preparation.



**Figure 8.** Absolute  $\zeta$  potential of  $\text{Al}_2\text{O}_3$ -water nanofluid after different durations of ultrasonication.



**Figure 9.** Viscosity of 0.5 vol %  $\text{Al}_2\text{O}_3$ -water nanofluid after different durations of ultrasonication.

fact that, without ultrasonication, the nanoparticles were not able to spread homogeneously in the base fluid and, therefore, a strong agglomeration occurred. Furthermore, this study started with the measurement of viscosity at 15 °C, and the particles could not get enough time to sediment, so the strong clusters of nanoparticles made resistance to the spindle, and viscosity increased. Therefore, from 15 to 25 °C, the viscosity of the nanofluids without ultrasonication dropped sharply from 1.58 to 1.40 mPa·s mainly because of two effects: One is the rotation

of the spindle of the rheometer, which was 100 rpm. It took about 10 min to change the temperature of the bath from 15 to 25 °C, and the spindle was rotating during this period. Therefore, some clusters broke and reduced the resistance to flow. The second reason is the temperature intensification, which is related to Brownian motion, and particles started to move from the cluster.

The relationship between the viscosities of 0.5 vol %  $\text{Al}_2\text{O}_3$ -water nanofluid with different ultrasonication durations is shown in Figure 9. Initially, the viscosity of the  $\text{Al}_2\text{O}_3$ -water nanofluid increased with increasing sonication time and reached a maximum value for an ultrasonication duration of 60 min; then, the viscosity of the nanofluid decreased with increasing sonication time, as shown in Figure 9. This trend is similar to the results obtained by Garg et al.,<sup>8</sup> who studied four different nanofluids consisting of 1 wt % MWCNTs in deionized water with GA. Ruan and Jacobi<sup>5</sup> also found the same viscosity trend for 0.5 wt % CNTs in EG with different ultrasonication durations. Moreover, they repeated the experiments at different shear rates, and the same results were observed. In addition, for further ultrasonication until 180 min, the viscosity of the  $\text{Al}_2\text{O}_3$ -water nanofluid decreased gradually, approaching the viscosity of the base fluid. The same trends were also found at different temperatures from 15 to 45 °C. It can be observed from Figure 9 that there are strong relationships between temperature and ultrasonication duration with the viscosity of the nanofluid. This phenomenon is also due to the effect of Brownian motion and van der Waals forces. At 15 and 25 °C, it can be seen that, after 120 min of ultrasonication, the viscosities are almost the same because the nanofluid became homogeneous after 120 min and there was no significant change in viscosity after extended durations of sonication. However, at 35 °C, the viscosity decreased slightly until 180 min of ultrasonication, and at 45 °C, the viscosity decreased significantly until 180 min of ultrasonication. Therefore, a nanofluid at higher temperature requires more energy to become well dispersed to achieve a lower viscosity. The TEM microstructures of Figures 3 and 5 also support the reason for the above phenomena.

The higher viscosity for an ultrasonication duration of 60 min in comparison with 0 and 30 min can be explained by the study of Starr et al.,<sup>20</sup> which claimed that a nanofluid with clustered nanoparticles exhibits a lower viscosity than a dispersed suspension. However, the amount, ratio, and

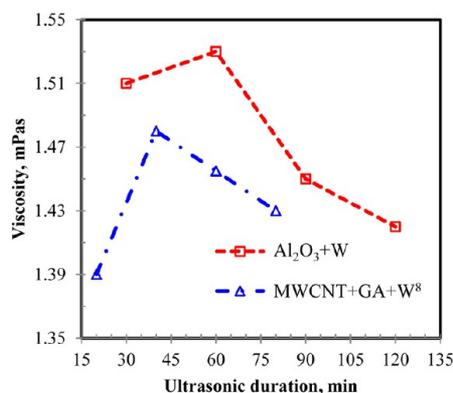
condition of clusters have significant effects on the viscosity of nanofluids. Here, in the cases without ultrasonication (i.e., 0 min) and with 30 min of ultrasonication, the nanoparticles were present in highly clustered forms (as seen in Figures 3 and 5), and because of self-weighting, they stayed at the bottom of the beaker. Therefore, this cluster has little effect on the flow of resistance as the viscometer spindle senses the resistance up to a certain level of suspension. That is why the viscosity was low compared to that at 60 min of ultrasonication. However, during 60 min of ultrasonication, the nanoparticle clusters started breaking into small clusters and reached the level of the sensing spindle. They were not taking part in the flow but rather creating resistance to flow, and viscosity was increased.<sup>21</sup> Because the surface-to-volume ratio was greater in the dispersed sample, the viscosity was higher as a result of increased attractive surface interactions. In addition, shorter ultrasonication duration refers to less dispersing energy. Thus, an ultrasonication duration of 30 min is not enough to provide the dispersing energy to overcome the agglomeration of nanoparticles, and the nanoparticles still remain in a clustered state. On the other hand, nanofluids with an ultrasonication duration of 60 min receive optimum energy to disperse the nanoparticles uniformly (as seen in Figures 3 and 5) and lose the agglomeration, thus resulting in higher viscosity. After 60 min of ultrasonication, it can be seen that the viscosity continuously decreased with further ultrasonication. Yang et al.<sup>14</sup> reported that a prolonged ultrasonication time affected the size and aspect ratio of particles, reducing the viscosity of the suspension. Therefore, in this experiment, it was observed that the viscosity of the nanofluid decreased after 60 min of ultrasonication. From the above discussion, two stages of morphological change were observed in the effect of ultrasonication duration on the viscosity of the nanofluids. Here, at the first stage, viscosity increases as ultrasonication loosens the agglomeration of the particles; however, in the second stage, successively prolonged ultrasonication broke up the particles' aspect ratio, and viscosity decreased.

The experimental results of this study were also compared with other published results. As the objective of this study was to determine the effect of ultrasonication duration on the viscosity of alumina–water nanofluid and there was no such information available for alumina–water nanofluid. Therefore, to confirm the trend of the viscosity variation with sonication time, the results of this study were compared with the results reported by Garg et al.<sup>8</sup> Figure 10 shows the effects of

ultrasonication duration on the viscosities of different nanofluids at 15 °C. The viscosity trend of Al<sub>2</sub>O<sub>3</sub>–water nanofluid with ultrasonication duration is similar to that of Garg et al.<sup>8</sup> In their research for ultrasonication duration from 20 to 80 min, viscosity initially increased from 20 to 40 min of ultrasonication and then decreased. It can be clearly seen from Figure 10 that maximum viscosity was found for an ultrasonication duration of 40 min. In this figure, the ultrasonication duration was considered from 30 to 120 min in lieu of considering the total ultrasonication duration of 0 to 180 min to make a good comparison with Garg et al.'s<sup>8</sup> results. The results of this current study show that the viscosity of the nanofluid increased up to an ultrasonication duration of 60 min and then decreased with increasing ultrasonication duration. The difference in viscosity level and the highest viscosity for an ultrasonication duration of this study with the other study is described below. The comparatively higher viscosity data of this study are due to the concentration variation, as the concentration was 0.5 vol % in this case and 0.1 wt % their case. Therefore, the highest viscosity data of this study were expected because of the higher concentration because viscosity increases with increasing particle concentration. The peak or highest viscosity for ultrasonication duration also depends on particle concentration. Based on the studies of Yu et al.,<sup>12</sup> a longer sonication time is required to disperse a higher amount of particles in solution. Thus, the highest viscosity value was found after 60 min of ultrasonication rather than 40 min of duration. Nevertheless, Ruan and Jacobi<sup>5</sup> also found the highest viscosity after 40 min of ultrasonication for 0.5 wt % MWCNTs in EG. However, the nanoparticles and base fluid were different, and there was the effect of a surfactant that was not considered. Furthermore, they studied the viscosity for some random ultrasonication durations such as 5, 40, 140, 520, and 1355 min. Other highest-viscosity points are likely to be found if other durations between 40 and 140 min are studied. As in this study, the gap between two consecutive ultrasonication durations was 30 min. If this gap were changed to 10 or 20 min, then the highest-viscosity position might change.

#### 4. CONCLUSIONS

The effects of ultrasonication duration (until 180 min) on colloidal structure and viscosity of 0.5 vol % Al<sub>2</sub>O<sub>3</sub>–water nanofluid were investigated. FESEM microstructure showed that nanoparticles were in aggregated form before being suspended in water. TEM analyses showed that nanoparticles start dispersing with ultrasonication and are properly dispersed in 90 min, after which they start to coalesce again, and after further sonication until 180 min, the nanoparticles are fully dispersed. The average particle and cluster size decreased with increasing ultrasonication duration. Stability and  $\zeta$  potential increased with sonication time. The present study has shown that there are strong relationships between the ultrasonication duration and the viscosity of nanofluid. The viscosity of Al<sub>2</sub>O<sub>3</sub>–water nanofluid decreased with increasing temperature. Moreover, the viscosity of the nanofluid increased to a maximum at an ultrasonication duration of 60 min and then decreased with increasing sonication time toward the viscosity of the base fluid. Therefore, throughout this study, it was found that the ultrasonication process changes the colloidal structure and affects the thermophysical properties of nanofluids. It can be concluded that, by using possible higher ultrasonication durations, more stable and lower-viscosity nanofluids can be obtained.



**Figure 10.** Variation of viscosity with ultrasonication duration at 15 °C.

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### Notes

The authors declare no competing financial interest.

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