



## Viscosity data for $\text{Al}_2\text{O}_3$ –water nanofluid—hysteresis: is heat transfer enhancement using nanofluids reliable?

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### Abstract

The effect due to temperature and particle volume concentration on the dynamic viscosity for the water– $\text{Al}_2\text{O}_3$  nanofluid has been experimentally investigated. The viscosity data were collected using a ‘piston-type’ commercial viscometer for temperatures ranging from room condition up to 75 °C. Two different particle sizes, namely 36 and 47 nm, have been considered. It has been found that, in general, nanofluid dynamic viscosity increases considerably with particle volume fraction but clearly decreases with a temperature increase. The viscosity values obtained for 36 and 47 nm particle-sizes are relatively close ones and others, except for high particle fractions. The complete viscosity database is presented. Results have clearly revealed the existence of a critical temperature beyond which the particle suspension properties seem to be drastically altered, which, in turn, has triggered a hysteresis phenomenon. Such a critical temperature has been found to be strongly dependent on both particle concentration and size. The hysteresis phenomenon has raised serious concerns regarding the reliability of using nanofluids for heat transfer enhancement purposes. Data have also shown that the Einstein’s formula and some other ones originated from the classical linear fluid theory seem to be limited to nanofluids with low particle fractions.

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### 1. Introduction

Nanofluid, a suspension of nanoparticles in a continuous and saturated liquid, has been found capable to provide a considerable heat transfer enhancement while compared to ‘conventional’ fluids such as water, ethylene glycol and engine oil. Some oxide nanoparticles seem to exhibit very good dispersion properties within liquids. In spite of their promising features, there are only few published results on nanofluids in confined flow situations—a review of relevant works may be found in Daughthongsuk and Wongwises [1] (see also Pak and Cho [2], Li and Xuan [3], Chein and Huang [4], Maïga et al. [5,6], Palm et al. [7] and Roy et al. [8]).

All of the research efforts were mostly focused on the characterization of nanofluid thermal/physical properties; a good proportion of these works was of the experimental nature and concerned with the measurement of effective thermal conductivity. A review of relevant works, see [9,10] in particular, has shown an important dispersion of the thermal conductivity data as obtained from various researchers. This dispersion is believed to be due to various factors such as the measuring techniques, the particle size and shape, the particle clustering and sedimentation. In spite of this, it was clearly found that the nanofluid thermal conductivity is well higher than that of the conventional heat transfer fluids [11–13]. Following the pioneering works by Masuda et al. [14], other relevant publications on nanofluid thermal conductivity include those by Choi [15], Pak and Cho [2], Lee et al. [16], Xuan and Li [17], Murshed et al. [9] and Liu et al. [18]. It is worth noting that there exist very few data dealing with the temperature effect on

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nanofluid thermal conductivity [14,19,20]. Most recently, the authors have performed measurement of the thermal conductivities for  $\text{Al}_2\text{O}_3$ –water nanofluid for particle concentrations as high as 9% [10]. Regarding the modeling of the nanofluid effective thermal conductivity, one should mention interesting models recently proposed by Koo and Kleinstreuer [21] and Chon et al. [22].

With regard to the nanofluid viscosity, the lack of data in the literature is even more striking. Masuda et al. [14] were the first who measured viscosity of some water-based nanofluids for temperatures varying from the ambient one to nearly 340 K, which was followed by Pak and Cho [2] who presented some additional data for  $\text{Al}_2\text{O}_3$ –water nanofluid. Wang et al. [11] have obtained, using three different dispersion techniques, some data for  $\text{Al}_2\text{O}_3$ –water and  $\text{Al}_2\text{O}_3$ –ethylene glycol mixtures at ambient temperature. Putra et al. [20] have provided some limited data showing the temperature effect on  $\text{Al}_2\text{O}_3$ –water nanofluid viscosity. Recently, Maré et al. [23] have obtained some new temperature-dependent viscosity data for  $\text{Al}_2\text{O}_3$ –water with relatively high particle concentrations. There exist, to our knowledge, no other data for the nanofluid dynamic viscosity, property that is of the crucial importance in every application involving with a fluid system.

In this work, we have performed extensive measurements of dynamic viscosity for the  $\text{Al}_2\text{O}_3$ –water nanofluid with two different particle sizes, 36 and 47 nm, and this for the temperature ranging from the room condition to nearly 75 °C.

## 2. Determination of particle suspension viscosity

From the theoretical point of view, a nanofluid represents a fascinating and interesting challenge to the researchers in the field of fluid dynamic and heat transfer, because it appears very difficult, if not to say quasi-impossible, to formulate any unified theory that can reasonably predict the nanofluid behaviour by considering it as a multi-component fluid. Since a nanofluid is also, by nature, a two-phase fluid, one may then expect that it would possess some common features with the solid–fluid mixtures. The question regarding the applicability and the limitation of the classical two-phase fluid theory for use for a nanofluid remains widely open.

For the determination of a particle suspension viscosity, there exist few theoretical formulas that can be used. It is very interesting to mention that almost all of the existing formulas were derived from the Einstein's pioneering work [24]. Based on the assumption of a linearly viscous fluid that contains dilute suspended spherical particles, he has calculated the energy dissipated by the fluid flow around a single particle and then, by associating that energy with the work done for moving this particle relatively to the surrounding fluid, he obtained:

$$\mu_r = \frac{\mu_{nf}}{\mu_{bf}} = (1 + 2.5\varphi) \quad (1)$$

where  $\varphi$  and  $\mu$  are, respectively, particle volume fraction and fluid dynamic viscosity; the subscripts bf, nf and r refer respectively to the base-fluid, the nanofluid and to the 'nanofluid-to-base fluid' ratio of viscosity. This famous formula was found

valid for a very low particle volume fraction, say 0.02 approximately (complete details and discussion may be found in Drew and Passman [25]). Since the publication of Einstein's work, there are many interesting and theoretical works all devoted to provide some 'corrections' to his formula. In these works, their authors have considered a negligible inertial effect in the fluid, which has rendered linear the equations of motion. A brief review of relevant works is given in the following (see also [25]).

Brinkman [26] has extended the Einstein's formula to a moderate particle volume concentration, say for concentration lower than 4%. His formula is as follows:

$$\frac{\mu_{nf}}{\mu_{bf}} = \frac{1}{(1 - \varphi)^{2.5}} \quad (2)$$

Frankel and Acrivos [27] have proposed:

$$\frac{\mu_{nf}}{\mu_{bf}} = \frac{9}{8} \cdot \left[ \frac{(\varphi/\varphi_m)^{1/3}}{1 - (\varphi/\varphi_m)^{1/3}} \right] \quad (3)$$

where  $\varphi_m$  is the maximum particle fraction that must be determined experimentally.

Lundgren [28] has proposed the following equation under the form of the Taylor series in  $\varphi$ :

$$\frac{\mu_{nf}}{\mu_{bf}} = \left[ 1 + 2.5\varphi + \frac{25}{4} \cdot \varphi^2 + O(\varphi^3) \right] \quad (4)$$

It is obvious that when the terms of second or higher order of  $\varphi$  are neglected, this formula is simply reduces to Einstein's one.

Batchelor [29] considered the effect due to the Brownian motion of particles for an isotropic suspension of rigid and spherical particles, and proposed:

$$\frac{\mu_{nf}}{\mu_{bf}} = (1 + 2.5\varphi + 6.5\varphi^2) \quad (5)$$

Graham [30] has proposed a generalization form of Eq. (3). His formula, which agrees well with Einstein's one for low value of  $\varphi$ , is as follows:

$$\frac{\mu_{nf}}{\mu_{bf}} = 1 + 2.5\varphi + 4.5 \times \left[ \frac{1}{\left(\frac{h}{d_p}\right) \cdot \left(2 + \frac{h}{d_p}\right) \cdot \left(1 + \frac{h}{d_p}\right)^2} \right] \quad (6)$$

where  $d_p$  and  $h$  are respectively the particle radius and inter-particle spacing.

It is apparent from the above formulas that the effective viscosity of a viscous fluid containing suspended solid particles is function only of the base fluid viscosity and the particle volume fraction. In principle, all of these formulas may be used for the determination of the nanofluid viscosity provided that the linear fluid assumption is satisfied. The limitation and the applicability of such a use are not yet determined.

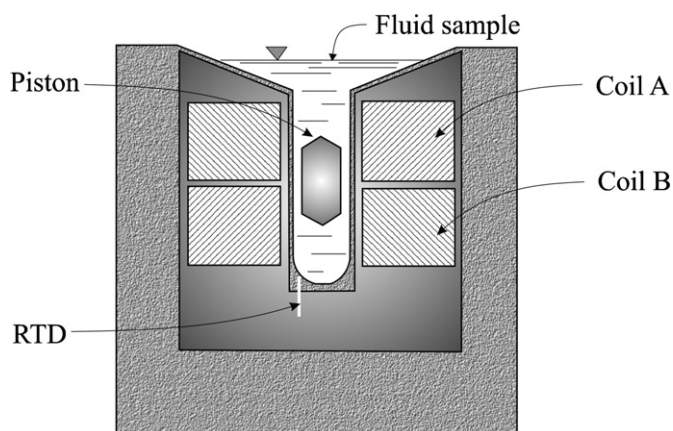
## 3. Experimental procedures

### 3.1. Description of instruments and apparatus

The apparatus used is mainly composed of a viscometer with a controller module and a heating jacket, Fig. 1(a). The viscometer, ViscoLab450 model from Cambridge Applied Systems, Massachusetts (USA), uses the 'piston-type' viscometer



(a)



(b)

Fig. 1. (a) ViscoLab450 and controller module. (b) Illustration of viscometer measurement chamber.

technology, Fig. 1(b). The measurement of viscosity is accomplished based on a Couette flow inside a cylindrical chamber; two magnetic coils installed inside a 316 stainless steel body are used to generate magnetically-induced forces on a cylindrical piston. The latter moves back and forth over a predetermined distance of 5 mm approximately. By alternatively powering the coils with a constant force, the total elapsed time corresponding to one round trip of the piston can then be measured, which, through a precise calibration process, is accurately related to the viscosity of the fluid sample. The back and forth movement of the piston creates a mixing effect inside the fluid sample, which ensures temperature uniformity of fluid inside the chamber. The fluid sample temperature is monitored using a precision Platinum RTD that is internally installed at the base of the chamber. Since fluid viscosity may significantly vary with temperature, it becomes important to know the exact temperature of the measurement chamber. The temperature accuracy and repeatability of the RTD probe are estimated to be  $\pm 0.2$  and  $\pm 0.1$  °C while the viscosity accuracy and repeatability are, respectively,  $\pm 1\%$  and  $\pm 0.8\%$ , accordingly to the manufacturer. The heating jacket is electrically heated by means of a standard 50 W nominal power cartridge heater. During the heating phase,

precautions were exercised in order to maintain the maximum temperature of the viscometer sensor around 85 °C as recommended by the manufacturer to avoid any possible damages to the internal wiring.

### 3.2. Preparation of nanofluids, experimental procedure and validation tests

In the present study, we are interested to establish a viscosity data base for  $\text{Al}_2\text{O}_3$ –water nanofluid with 36 and 47 nm average particle diameters. These mixtures have been purchased readily prepared and mixed (Nanophase Technologies, USA). At delivery, they had, respectively, as particle volume fractions, approximately 22 and 15%. In order to create other particle volume concentrations, a diluting process with distilled water followed by a vigorous mechanical stirring action was found to be sufficient. It should be noted that since some dispersing agents were used, the suspension stability of nanoparticles within the base fluid, distilled water, has been found to be very good even after a relatively long resting period, say for weeks or months. Several particle volume concentrations varying from 1 to nearly 13% have been obtained.

The experimental procedure is quite simple. At the beginning of an experiment, the piston is first removed from the measurement chamber; the latter is then half filled with the fluid (about 10 ml). The piston is next transferred back into the chamber, which is then full filled with the fluid. With the steel jacket readily mounted on the outside of the sensor, the heating circuit and the viscometer controller module are set on. A reading of viscosity was taken only once the viscosity/temperature data are stabilized. For a typical experiment, using the full power of the cartridge heater (50 W), the necessary duration of the heating phase has been found to be approximately 4 h in order to raise the fluid sample temperature up to nearly 80 °C from the ambient condition. In conjunction with a very low mass of fluid sample, such a long heating duration has been found necessary to ensure a sufficiently low temperature gradient in order to minimize any lag, if any, between the temperature reading and viscosity measurement. Because of such a long elapsed-time, it has been necessary to perform, at regular time interval, a forcing mixing action within the fluid sample inside the measurement chamber, using a ‘purging feature’ (rapid strokes of the piston) thus avoiding deposition of nanoparticles. In this study, we were also interested to determine whether there exists some hysteresis behavior due to the heating process on particle suspension quality. In order to realize such a study, after reaching a maximum temperature, the heating power is turned off thus allowing the system to be cooled by natural convection. During such a cooling phase that also took 5 h approximately to reach the ambient temperature, continuous readings of temperature and viscosity were carried on.

In order to verify the viscometer accuracy as well as to assess the reliability of the experimental procedures, two different sets of viscosity measurement were carried out. The first set of tests has been conducted using CAS calibration fluid (a mineral oil with the code name S3S) that was supplied by the manufacturer, for which viscosity data were available. For the second set

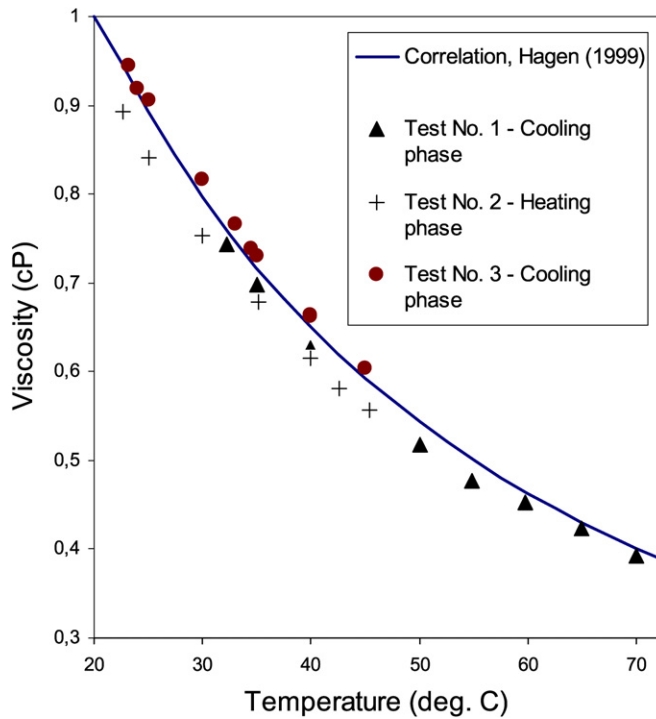


Fig. 2. Results from validation tests using distilled water.

of validation tests, distilled water has been used. For both fluids, values of viscosity have been collected during both the heating and cooling phases. Measured viscosity values of CAS fluid have been found to be very close to the tabulated ones: in fact, the maximum relative error does not exceed, so far, 3%, for temperatures ranging from 20 to nearly 45 °C [31]. Results from the validation tests using distilled water, Fig. 2, have clearly shown that the viscometer also performs quite satisfactorily: in fact, the maximum relative error between the measured viscosity values and the corresponding tabulated ones did not exceed, in any case, 6.5%. It is worth noting that the following correlation [32] has been employed for computing water dynamic viscosity:

$$\mu_{bf} \times 10^4 = e^{(1.12646 - 0.039638 \cdot T) / (1 - 0.00729769 \cdot T)} \quad (7)$$

where  $T$  (K) is temperature and  $\mu$  in cP. Therefore, ones can conclude with confidence about the viscometer reliability and also about the appropriateness of the experimental procedures adopted.

#### 4. Presentation of viscosity data

Fig. 3 shows the results for the relative viscosity (defined as the ratio of ‘nanofluid/water’ viscosity) as obtained for  $\text{Al}_2\text{O}_3$ –water under the ambient condition and particle volume fraction varying from 0.15% to a value as high as 13%. Some other data are also shown for discussion. It is worth mentioning that based on the satisfactory validation tests using calibration fluid and water one can expect a maximum relative error of  $\approx 6.5\%$  for the measured values of viscosity.

As we may expect, the nanofluid effective viscosity clearly increases with increasing particle concentration. Thus, for

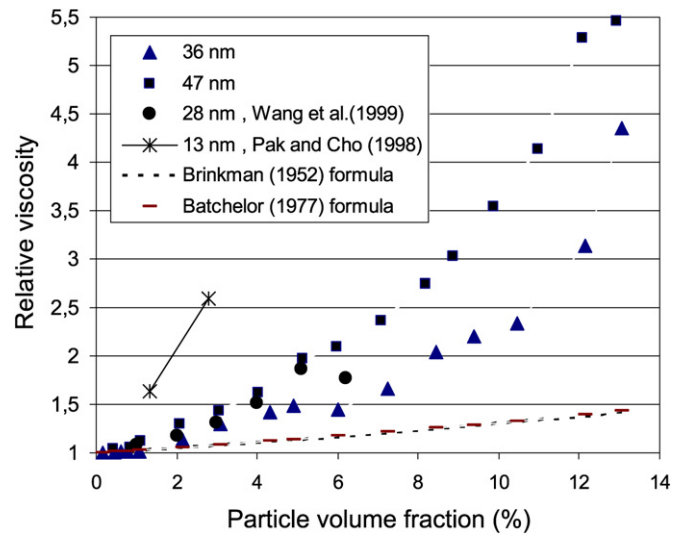


Fig. 3. Water– $\text{Al}_2\text{O}_3$  relative viscosity values at room condition.

47 nm-alumina–water for example, the relative viscosity has increased from  $\approx 1.12$  to  $\approx 1.6$ , to  $\approx 3.0$  and then to  $\approx 5.3$  for particle fraction increasing from 1 to 4%, to 9% and then to 12%. Such a behavior was also observed for 36 nm particle-size nanofluid: the relative viscosity has increased from  $\approx 1.1$  to  $\approx 1.4$ , then to  $\approx 2.0$  and finally to  $\approx 3.1$ , respectively, for particle fractions 2.1, 4.3, 8.5 and 12.2%. It is very interesting to observe that the viscosity values corresponding to 36 nm particle-size are lower than those with 47 nm particle-size. Such a difference clearly becomes more pronounced for a particle fraction higher than 5%.

The results in Fig. 3 have obviously shown that both formulas proposed by Brinkman [26] and Batchelor [29] underestimate severely nanofluid viscosity, except for a very low particle fraction (lower than 1%). Furthermore, one may notice an apparent contradictory behavior from Fig. 3 regarding the particle-size effect. In fact, Pak and Cho [2] data obtained for 13 nm size are well higher than all other data. On the other hand, Wang et al. [11] data obtained for 28 nm particle-size fall between the present data for 36 and 47 nm. It appears difficult to draw a conclusive statement about such a result, which may be due to various factors such as the methods used for nanofluid preparation as well as the differences in the technique of measurement. As stated by Koblinski et al. [33], this is in fact one of the major concerns regarding the nanofluid study. For computing purpose, the following correlations have been proposed, respectively, for 47 and 36 nm particle-sizes:

$$\mu_r = \frac{\mu_{nf}}{\mu_{bf}} = 0.904e^{0.1483 \cdot \varphi} \quad (8)$$

$$\mu_r = (1 + 0.025 \cdot \varphi + 0.015 \cdot \varphi^2) \quad (9)$$

##### 4.1. Temperature-dependent viscosity data

A large number of viscosity values were collected for the  $\text{Al}_2\text{O}_3$ –water nanofluid, two different particle-sizes, 36 and 47 nm, particle volume fraction ranging from 1 to nearly 9.4% and temperature ranging from 21 to 75 °C approximately. It is



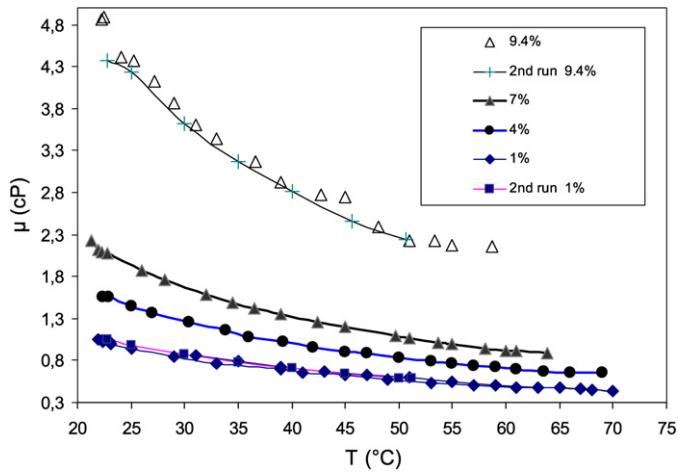
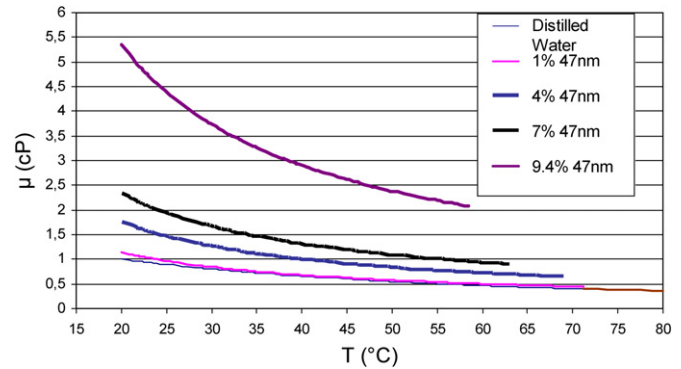


Fig. 4. Viscosity data for water–Al<sub>2</sub>O<sub>3</sub>–47 nm and particle volume fractions ranging from 1 to 9.4%.

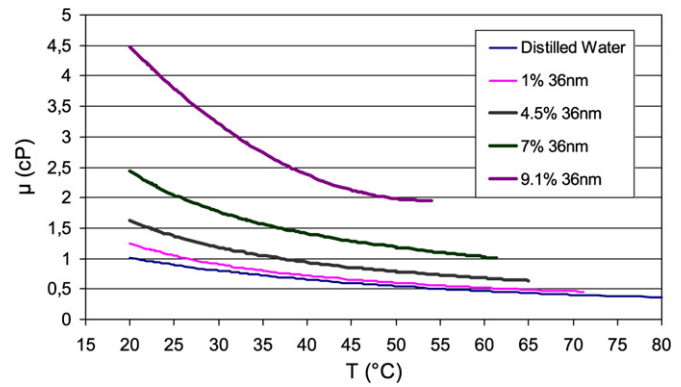
worth noting that for given nanofluid and particle volume fraction, at least two different tests were performed using fresh fluid samples each time, in order to ensure the consistency as well as the repeatability of the measured data.

It has been observed that, in general, nanofluid viscosity considerably increases with increasing particle volume fraction, but clearly decreases with an increase in temperature. The effect due to the particle fraction is linked to the fact that increasing concentration would have a directly influence on internal viscous shear stresses. On the other hand, an increase of fluid temperature has obviously a weakening effect on the inter-particle/inter-molecular forces. Thus, for water–Al<sub>2</sub>O<sub>3</sub> and 47 nm particle-size in particular, viscosity values at 30 °C are approximately 0.8, 1.4, 1.7 and 3.6 cP, respectively, for volume concentrations of 1, 4, 7 and 9.4%, Fig. 4. Similar behavior has also been observed for the nanofluid with 36 nm particle-size. It is very interesting to observe that for the nanofluid under consideration, the temperature gradient of viscosity is generally more pronounced for temperatures around the ambient condition, say for the range 22 to 40 °C. Such a gradient also appears higher for high particle fraction, the case 9% for example. This result strongly suggests that the temperature effects on the particle suspension properties may be very different for high particle fraction than for a lower one. With an increase of temperature, it has been observed that viscosity tends to become nearly constant independently of temperature. In fact, for a given particle fraction, it has been clearly observed that there exists a critical temperature,  $T_{cr}$ , beyond which irreversible damages seem to be produced to the particle suspension properties. This has resulted in a rather erratic increase of the nanofluid viscosity, behavior that remains not very well understood and is presented in details in Section 4.2. For a given concentration,  $T_{cr}$  corresponds to last data point i.e. the one with the highest temperature on corresponding curve (Fig. 4).

Figs. 5(a) and 5(b) show, respectively, the complete tendency curves of nanofluid viscosity as obtained for the 47 and 36 nm particle sizes. It is worth mentioning that for each of the figures, the lowest curve presents distilled water viscosity as given by Eq. (7) while the other ones, from the second lowest to the high-



(a)



(b)

Fig. 5. Viscosity tendency curves for: (a) Water–Al<sub>2</sub>O<sub>3</sub>–47 nm, particle volume fractions of 1, 4, 7 and 9.4%. (b) Water–Al<sub>2</sub>O<sub>3</sub>–36 nm, particle volume fractions of 1, 4.5, 7 and 9.1%.

est one, correspond to the other four different particle fractions tested. A similar behavior regarding the temperature and particle concentration effects on viscosity can be again observed in these figures. As far as  $T_{cr}$  is concerned, it is simply given by the highest temperature point corresponding to the end of each of the curves shown.

In this work, the authors were particularly interested to determine whether the formulas presented earlier in Section 2 can be applied to the present nanofluids. From the viscosity data collected, the relative viscosity values have been calculated and expressed as a function of the temperature and particle fraction. Results have shown that for a given particle fraction that is lower than 4%, the values for the relative viscosity are approximately identical for the particle-sizes studied and remain constant independently of temperature. For a higher particle fraction, say for 7 and 9%, it has been found that the relative viscosity is clearly dependent not only on the temperature but also on the particle-size as well. As a consequence, it has been determined that none of formulas cited would be appropriately used for the nanofluids considered (Desgranges [34]). In fact, even for a relatively low particle fraction, the Einstein's formula and the ones proposed by Brinkman [26], Lundgren [28] and Batchelor [29] have all underestimated nanofluid viscosity. Thus, for the particle volume fraction of 1% in particular, these formulas have predicted 1.025–1.026 range for relative viscosity, while the corresponding values from our data fall within the interval 1.05–1.29. For the case of 4% fraction, the relative vis-

cosity values are within the interval 1.1–1.11, as computed by these formulas, values that are drastically underestimated while compared to our results, 1.43–1.73. The main reason for such a discrepancy resides in the fact that the Einstein's formula as well as the other ones originated from it have been obtained based on the assumption of a linear fluid surrounding an isolated particle, situation that appears to be far from the case of nanofluid. Therefore, the following correlations have been proposed for use with the alumina-water nanofluids and particle volume fractions of 1 and 4%, respectively:

$$\mu_r = 1.125 - 0.0007 \cdot T \quad (10)$$

$$\mu_r = 2.1275 - 0.0215 \cdot T + 0.0002 \cdot T^2 \quad (11)$$

where  $T$  (°C) is fluid temperature. On the relative basis, the above correlations exhibit, respectively, average errors of 0.06 and 1.28%, and standard-deviations of 3.75 and 11.39%, values that are believed very acceptable in conjunction with the current experimental uncertainty. Unfortunately, for higher particle volume fraction, it was not possible to provide correlation that can reasonably take into account the combined temperature, particle concentration and size effects.

#### 4.2. Hysteresis phenomenon on viscosity

As mentioned earlier, our experimental data have clearly revealed that for a given particle volume concentration, there is a critical temperature beyond which nanofluid viscous behavior becomes drastically altered. In fact, if the fluid sample is heated beyond such a critical temperature, a striking increase of viscosity occurs. And if it is allowed to cool after being heated beyond a critical temperature, then a hysteresis phenomenon can occur. Such an interesting behavior can be better understood by scrutinizing Fig. 6, which shows the viscosity data as obtained for the nanofluid with 47 nm particle-size and a particular particle volume fraction of 7%. First, the Run 1 (heating phase) has been executed that is immediately followed by a cooling phase identified as Run 1a, Fig. 6. The viscosity data obtained from the Run 1 have shown that at the temperature of 61.3 °C approximately, the nanofluid viscosity has reached its lowest level, and beyond 61.3 °C, the viscosity starts to increase with

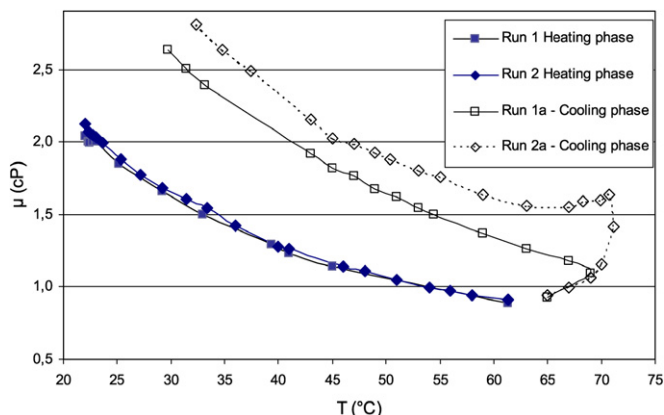


Fig. 6. Hysteresis observed for water–Al<sub>2</sub>O<sub>3</sub>–47 nm, 7% particle volume fraction.

an increase in temperature. Such a behavior has been confirmed by performing another run, the Run 2 (heating phase). One can notice here a very low dispersion of the data collected from Runs 1 and 2, which has assessed with confidence not only the data consistence but also the reliability of the instruments used and the experimental procedures adopted. Now, as the fluid sample is heated beyond 61.3 °C and being let cooled slowly afterwards, an interesting hysteresis behavior on viscosity has been observed. Thus, during the two different cooling schemes performed, Runs 1a and 2a, the measured values of nanofluid viscosity for a given temperature are remarkably higher than those collected during the heating phase (Runs 1 and 2). One can then expect that during the Runs 1a and 2a, there would be some irreversible damages that have been done to the particle suspension properties. It is worth noting that the only difference between the Run 1a and Run 2a resides in the value of maximum temperature reached by the fluid sample prior to be cooled: this temperature was approximately 69 °C for Run 1a and 71.2 °C for Run 2a. One can also observe in Fig. 6 that the viscosity values for Run 2a are clearly higher than those of Run 1a. Such a result likely indicates that the damages to the particle suspension properties are more pronounced in the case of Run 2a. Thus, the temperature level to which the fluid is heated up has a great importance on the possible impact on the nanofluid viscous behavior and suspension properties. It is very interesting to mention that this intriguing hysteresis behavior has also been observed for other particle fractions, for Al<sub>2</sub>O<sub>3</sub>–water with 36 nm particle-size as well as for CuO–water nanofluid with 29 nm particle-size.

Fig. 7 shows another striking example of the hysteresis behavior, which has been observed for the nanofluid with 36 nm particle-size and four particle volume concentrations. It is very interesting to note that for a low particle volume fraction, say 1% in particular, the hysteresis behavior was not observed for both the particle-sizes tested. However, for a higher particle fraction, this hysteresis does exist and clearly becomes more pronounced with an increase of the particle volume fraction. Thus, for 7 and 9% particle fractions for example, such a hysteresis behavior was rather severe. It is worth mentioning that for the 9% particle fraction in particular, the heating phase be-

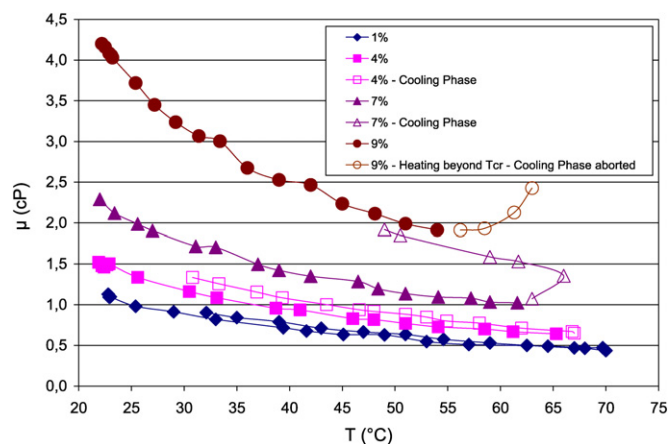


Fig. 7. Hysteresis observed for water–Al<sub>2</sub>O<sub>3</sub>–36 nm, particle volume fractions of 1, 4, 7 and 9%.

Table 1  
Values of  $T_{cr}$  (°C) for the nanofluids studied

	Al <sub>2</sub> O <sub>3</sub>		CuO
	47 nm	36 nm	29 nm
1%	N/O*	N/O*	66.2
≈4%	69	65.3	63.2
7%	63.8	61.6	57.2
≈9%	58.7	54	51

Note. N/O\* Hysteresis phenomenon not observed for the temperature range considered.

yond the critical temperature was aborted because the fluid has become very viscous in such a way that the viscometer piston has been stuck inside the measuring chamber, which obviously indicates a rather drastic alteration of the nanofluid rheological properties.

Furthermore, it has been clearly observed that when a heated fluid sample is maintained below the corresponding critical temperature prior to be cooled, the hysteresis phenomenon did not occur. Table 1 presents finally the values for the critical temperature as experimentally determined for the three nanofluids studied. It has been observed that the critical temperature appears strongly dependent of both the particle concentration and size. In fact,  $T_{cr}$  has been found to decrease with an increase of the particle fraction for a given particle size. On the other hand, for a given particle concentration,  $T_{cr}$  clearly decreases for a smaller particle size.

Such an intriguing hysteresis phenomenon still remains not very well understood. It has been suggested that the remarkable increase of nanofluid viscosity occurred during the cooling schemes 1a and 2a, see again Fig. 6, may be a result of some drastic yet unknown changes that have been produced into the nanofluid under the temperature effect. In fact, a visual observation of the fluid samples at the end of the Runs 1a and 2a has clearly shown an appearance of a highly viscous fluid with a visible sign of particle agglomeration on the measuring chamber inner surface. Such observations obviously indicate that the particle suspension properties have tremendously been altered or even destroyed when the fluid sample is heated beyond the critical temperature. This rather interesting fact may be closely linked to the presence of some dispersants i.e. chemical substances often used as surfactants necessary to maintain the particles in a stable suspension. Ones can believe that beyond a critical temperature, the expected effects from these dispersants may be greatly reduced or even destroyed, which causes the particles to lose their suspension capabilities. They have then the tendency to agglomerate that, in turn, results in an erratic and drastic increase of the nanofluid viscosity as it has been shown earlier. Such statements appear quite plausible as they can explain that once the deterioration of the particle suspension properties is initiated, a presence of more particles inside the base fluid—i.e. in the cases of a higher particle volume fractions for a given particle-size/or a smaller particle-size for a given particle volume fraction—would logically result in a more pronounced effect on the nanofluid viscous behavior. Such an explanation seems to be well corroborated with the values of  $T_{cr}$  shown in Table 1 earlier. Unfortunately, it was not

possible to us to provide any information regarding the nature and the characteristics of the dispersants that were used by the manufacturer, in order to verify such arguments. Hence, more investigations will be needed to completely understand such an interesting phenomenon.

As it has been mentioned previously regarding the lack of thermal property data for the nanofluid, the viscosity database presented in this work, which is believed among the first of a kind, constitutes an interesting contribution towards a complete understanding of those special fluids. Finally, it is worth mentioning that the hysteresis phenomenon observed in this work has not yet been reported in the literature.

#### 4.3. Hysteresis: is heat transfer enhancement using nanofluid reliable?

In light of the hysteresis phenomenon, one can logically wonder whether it will be problematic to consider using nanofluids for heat transfer enhancement purpose. It must be quite clear at the present stage that in an eventuality that such a hysteresis behavior of viscosity occurs for a nanofluid flow, then the entire flow and thermal field and the heat transfer behavior could be, as a direct consequence, erratic and unpredictable as well because of the local changes in fluid viscosity. For a non-isothermal flow situation, the behavior and development of both the flow and thermal field would become highly unpredictable. Another direct and important result of this resides in the fact that the heat transfer would no longer be appropriately determined. Such a scenario, if it does occur, will be rather unfavorable and can seriously compromise the use of a nanofluid in thermal applications.

It is important to recall, however, that for some confined flow situations, many researchers have clearly established experimentally that an inclusion of the nanoparticles within a viscous fluid has produced a substantial and interesting heat transfer enhancement [2,3,35]. In these works, the hysteresis phenomenon was not observed probably because of a relatively low temperature range considered. On the other hand, it is also plausible that the presence of a strong forced convection flow may possibly mask the effect, if any, which would result from a hysteresis behavior. Nevertheless, based on the above experimental evidence, one can state with confidence about the heat transfer enhancement capability of the nanofluids for a forced convection flow and this, in spite of the fact that the physical mechanism behind such an enhancement has not yet been completely understood to date [33]. On the other hand, it has also been found, through experimental observations and data, that a high temperature may drastically alter or even destroy the particle suspension properties, which in turn, would have a direct impact on the heat transfer. In fact, it has been observed that for the cases of a natural convection flow inside an enclosure as well as in a case of pool boiling using Al<sub>2</sub>O<sub>3</sub>–water nanofluid, such an alteration of the particle suspension properties has been clearly found to be responsible for a deterioration of the surface heat transfer coefficient (see in particular, Putra et al. [20] and Nguyen et al. [36]).

The obvious question is then how to produce a ‘right’ nanofluid in order to accomplish the heat transfer enhancement purpose. In other words, in light of the hysteresis phenomenon shown herein, one may wonder whether it is possible to properly produce a nanofluid in such a way that the good i.e. stable particle suspension can be maintained for a wide range of temperature. Such a formidable task represents, in itself, a rather interesting challenge from the manufacturing standpoint. In fact, it has been known that the particle suspension properties are closely influenced not only by the properties of the constituents used but also by the preparation methods adopted to produce the suspension itself.

Finally, it is worth mentioning that the nanofluid behavior and characteristics are merely known in a real thermal application [33], and more research works will, indeed, be necessary in the future.

## 5. Conclusion

In this work, the new and complete viscosity database has been established for the water– $\text{Al}_2\text{O}_3$  nanofluid with 36 and 47 nm particle sizes. The viscosity data have been obtained for the ambient condition and particle volume fraction varying from 1 to nearly 13%. The temperature and particle-size effects have been thoroughly investigated, considering temperature ranging from 22 to 75 °C and particle volume fraction from 1 to 9.4%. It has been found that in general, the nanofluid viscosity strongly depends on both temperature and concentration; while the particle-size effect seems to be important only for sufficiently high particle fraction. The existence of a critical temperature has clearly been established. Beyond such a critical temperature, the particle suspension properties seem to be irreversibly altered, which, in turn, may induce a hysteresis behavior. The critical temperature has been found strongly dependent on both particle fraction and size. The hysteresis phenomenon observed on viscosity has raised serious concerns regarding the use of nanofluids for heat transfer enhancement purposes. Several correlations are provided to compute nanofluids viscosity. Finally, it has been found that the Einstein’s formula and its derived ones that were originated from the classical theory of linear fluid appear to be limited to nanofluids with low particle volume fractions.

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