Hysteresis Phenomenon on Water-Based Nanofluid Viscosity

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Abstract: - Dynamic viscosity values for three particular water-based nanofluids have been experimental determined using a commercial 'piston-type' viscometer. Viscosity data, expressed as a function of temperature and particle volume fraction, are presented for 29nm particle-size CuO-water and Al_2O_3-water with 36nm and 47nm particle-sizes, temperatures up to 75°C and particle volume fraction up to 9%. In general, the nanofluid viscosity increases considerably with an increase of particle volume fraction but decreases with increasing temperature. The existence of a critical temperature has been clearly established, beyond which the hysteresis behaviour on viscosity has been observed. Such a critical temperature has been found strongly dependent on both particle volume fraction and size. This hysteresis phenomenon has raised major concerns regarding the use of nanofluids for the heat transfer enhancement purposes.

Key-Words: - Water-based nanofluids, Al_2O_3-Water, CuO-Water, Alumina nanoparticles, Copper-oxide nanoparticles, Experimental study, Dynamic viscosity, Hysteresis.

1   Introduction

A nanofluid, a mixture of nanoparticles in a continuous and saturated liquid, has been found capable to provide a considerable heat transfer enhancement while compared to the 'conventional' fluids such as water, ethylene glycol and engine oils. It is worth noting that some oxide particles exhibit good dispersion property in water. There are, unfortunately, few published results and data on the nanofluids for use in a confined flow configuration – a review of relevant works may be found in [1], see also [2-7]. The previous studies were mostly concerned with the characterization of the nanofluid thermal and physical properties, among them, a good portion was of the experimental nature and shown the data for the effective thermal conductivity. A review of relevant literature [8, 9] has shown, however, an important dispersion of data, which may be due to various factors such as the measuring techniques, the particle size and clustering and also particle sedimentation. In spite of this, it has clearly been found that the nanofluid thermal conductivity is well higher than that of the base fluid [10-12]. Parting from Masuda and colleagues' pioneering work [13], one can cite several other relevant publications on the nanofluid thermal conductivity, see [2, 8, 14-17]. Furthermore, it is important to note that there very are few data considering the

temperature effect on nanofluid thermal conductivity [13, 18, 19]. Recently, the authors have obtained some new thermal conductivity data the Al_2O_3-water nanofluid with particle fraction as high as 9% [9].

With regard to the nanofluid viscosity, the lack of data is even more striking. Masuda et al. [13] were likely the first to obtain the viscosity data for several nanofluids and temperatures varying from the ambient condition to nearly 340K. Pak and Cho [2] followed with their data obtained for Al_2O_3-water nanofluid and two particle volume fractions. Wang et al. [10] obtained limited data for Al_2O_3-water and Al_2O_3-ethylene glycol at ambient temperature. Putra et al. [19] have provided few data showing the temperature effect on Al_2O_3-water nanofluid viscosity. Most recently, Maré et al. [20] obtained some temperature-dependent viscosity data for Al_2O_3-water with a relatively high particle fraction. There are, to our knowledge, no other data for the nanofluid viscosity, property of highly importance for every thermal application that uses a fluidic system.

In this work, an extensive viscosity measurement has been carried out for three particular water-based nanofluids, CuO-water with 29nm particle-size and Al_2O_3-water with two different particle-sizes, 36nm and 47nm, and this for temperatures varying from the room condition to

nearly 75ºC. Some data are presented with an emphasis on the hysteresis phenomenon that has been experimentally observed while heating the fluid samples beyond a critical temperature.

2   Instruments and Experimental Procedures

The apparatus consists of a viscometer, a controller module and a heating jacket, Fig 1a. The viscometer, ViscoLab450, from Cambridge Applied Systems, Massachusetts (USA), uses the 'piston-type' technology, Fig. 1b. The functioning of such a viscometer is as follows: two magnetic coils installed inside a stainless steel body are used to generate a magnetically-induced force on a piston. The piston moves back and forth over a predetermined distance of ≈5mm. By alternatively powering the coils with a constant force, the total elapsed time corresponding to a round trip of the piston can then be measured, which, through a precise calibration process, is accurately related to the fluid viscosity. The fluid sample temperature is monitored using a precision Platinum RTD internally installed at the chamber base. The accuracy and repeatability of such a RTD probe are estimated ±0.2ºC and ±0.1ºC, while the viscometer accuracy and repeatability are ±1% and ±0.8% accordingly to the manufacturer. The steel heating jacket is electrically heated by means of a standard 50W nominal power cartridge heater. During the experiments, a precaution was constantly exercised to ensure that the viscometer maximum temperature did not exceed 100ºC in any case to avoid possible damages to the internal wiring.

In the present study, we are interested to establish the viscosity database for three particular water-based nanofluids, namely Al_2O_3-water nanofluid with 36nm and 47nm average particle-diameters, and CuO-water with 29nm particle-size. These mixtures have been purchased readily prepared and mixed (Nanophase Technologies, USA). At delivery, they had as original particle volume fractions, approximately 22%, 17% and 14% for the nanofluids cited earlier. In order to produce other particle volume fractions, a proper diluting process using distilled water followed by a rigorous stirring action has been found necessary. It should be noted that as some dispersing agents were used by the manufacturer, the particle suspension stability within water has been found to be quite acceptable even for a relatively long resting period. Several particle volume fractions ranging from 1% to nearly 13% have been obtained.

The experimental procedure is simple. Starting an experiment, the piston is first removed from the chamber, which is then half filled with a
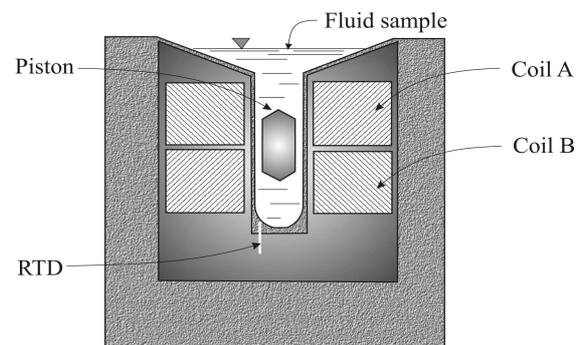
Fig. 1 Viscometer with controller and heating jacket

volume of fluid (about 5ml). The piston is next transferred back to the chamber that is then full filled with fluid sample. The heating is set on. In general, a reading of the viscosity can only be taken once the viscosity/temperature data are stabilized. For a typical experiment, the heating phase duration is approximately 4hr to raise the fluid sample temperature up to nearly 80ºC from the ambient condition. Such a long duration was necessary to ensure to minimize any lag between temperature reading and viscosity measurement. In this study, we were also interested to determine whether there exists some hystereris behaviour due to the heating process on particle suspension quality. Thus, after reaching a maximum temperature, the heating is turned off that allow the entire system to be naturally cooled off by natural convection. During such a cooling that took nearly 5hr, continuous reading of fluid temperature and viscosity was continued.

In order to verify the viscometer precision and to assess the reliability of the experimental procedures, two different sets tests were performed. The first set of validation tests has been conducted using the mineral calibration oil supplied by the manufacturer. For the second set of tests, distilled water was used. For both fluids, the viscosity values were collected during both the heating and cooling

phases. The data collected using the calibration fluid for the temperature range 20ºC-45ºC have been found very close, within ±3%, with respect to the tabulated ones. For the case of water, the agreement was also found very acceptable while comparing the measured values and the tabulated ones. In fact, the maximum relative error did not exceed, so far, 6.5%, value that clearly confirms not only the satisfactory performance of the viscometer itself but also the reliability of the experimental procedures adopted.

3   Results and Discussion

3.1   Dynamic Viscosity Database

An extensive measurement program of viscosity was carried out for Al_2O_3-water nanofluid with averaged particle-sizes of 36nm and 47nm, particle volume fractions varying from 1% to nearly 9.4% and temperatures ranging from 21°C to 75°C approximately. It is worth noting that most data were collected from two different runs to ensure not only the consistence but the repeatability of the data as well. Such qualities can be noticed through the small data dispersion shown in Figure 2.

It is observed that, in general, nanofluid viscosity considerably increases with increasing particle volume fraction, but clearly decreases with increasing temperature. The effect due to the particle fraction is linked to the fact that increasing fraction would have a directly influence on the internal viscous shear stresses; while the temperature effect is obviously due to the weakening of inter-particle and inter-molecular forces. Thus, for water-Al_2O_3 and 47nm particle-size in particular, viscosity values at 30°C are approximately 0.8, 1.4, 1.7 and 3.6cP for particle fractions of 1%, 4%, 7% and 9.4%, respectively, Fig. 2. Similar behaviours were also observed for water-Al_2O_3 nanofluid with particle average-diameter of 36nm as well as for water-CuO nanofluid with 29nm particle-size. It is very interesting to observe that for the three nanofluids under study, the temperature gradient of viscosity is generally more important for temperatures near the room condition, say for temperatures from 22°C to 40°C approximately. Such gradient also appears particularly pronounced for sufficiently high particle volume fractions. This result strongly suggests that the temperature effect on the particle suspension properties may be very different for high particle fraction than for a lower one. With an increase of the temperature, it has been observed that the values of nanofluid viscosity tend to

become nearly constant regardless of temperature. In fact, for a given fraction, it was clearly observed that there is a critical temperature, T_{cr}, beyond which drastic and irreversible damages seem to be produced to the particle suspension properties. This has resulted in a rather erratic increase of the nanofluid viscosity. Such an intriguing behaviour, which has not yet been observed for the nanofluids, still remains not very well understood.
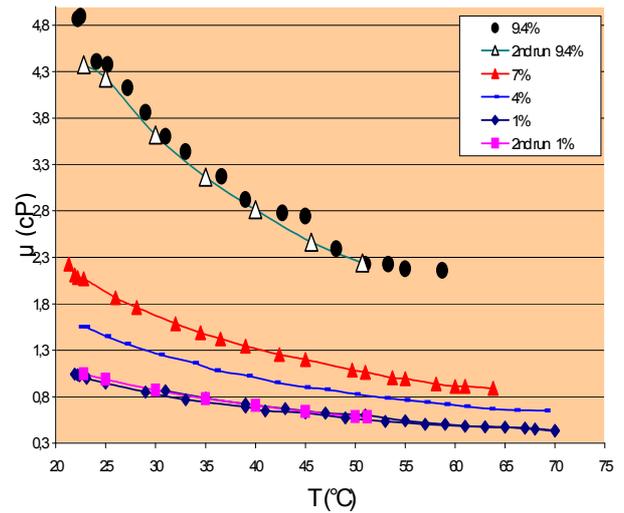
Fig. 2 Viscosity data for Water-Al_2O_3 with 47nm particle-size
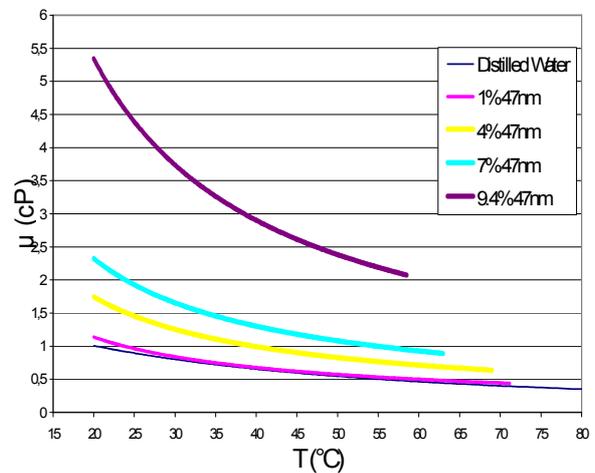
Fig. 3 Viscosity curves for Water-Al_2O_3 with 47nm particle-size

Figures 3-5 show the complete viscosity databases, represented by the continuous curves obtained from a least-square fitting of measured data for the three nanofluids studied. It is worth to mention that for each of these figures, the lowest curve corresponds in fact to distilled water while the other ones, from the second lowest to the highest one, correspond to the four particle fractions tested. The previously

discussed behaviours regarding the temperature-and-particle fraction effects can be clearly observed here again. The values of T_{cr} are simply given by the highest temperature values points of these curves.
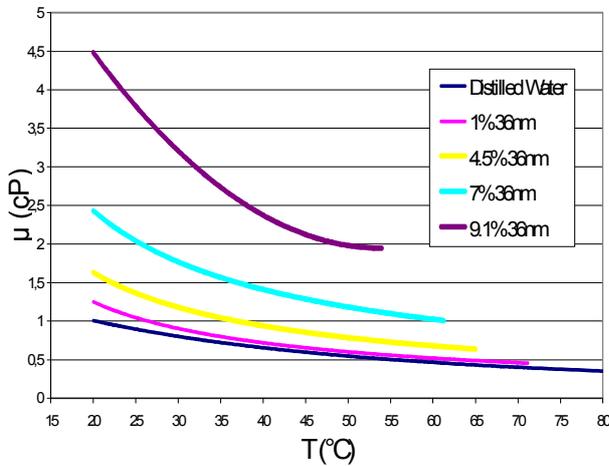
Fig. 4 Viscosity curves for Water-Al_2O_3 with 36nm particle-size
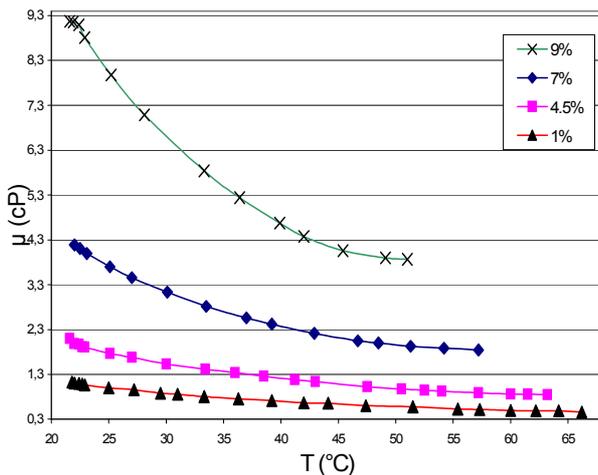
Fig. 5 Viscosity curves for Water-CuO with 29nm particle-size

3.2 Hysteresis Phenomenon On Viscosity

As mentioned earlier, our experimental data have clearly revealed that for a given particle volume fraction, it does exist a critical temperature beyond which the nanofluid viscous behaviour becomes profoundly altered. In fact, we observed that if the fluid sample is heated beyond such a critical temperature, a clear increase of nanofluid viscosity did result. And if the fluid sample is cooled after being heated beyond a critical temperature, then a hysteresis phenomenon can occur. Such an intriguing phenomenon is better understood by scrutinizing Fig. 6 that presents viscosity data

obtained for 47nm particle-size, 7% particle volume fraction subject to two different cooling schemes.
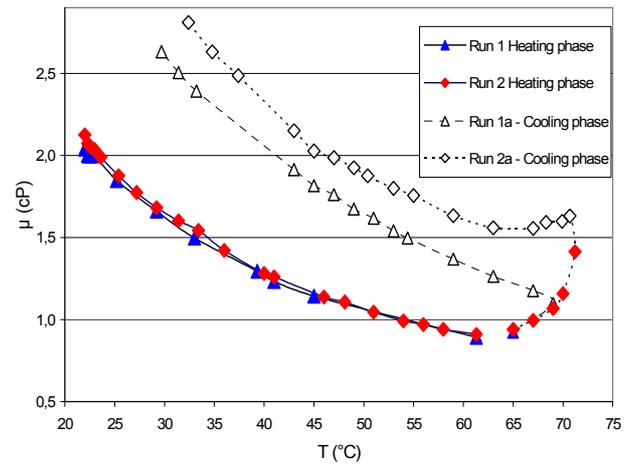
Fig. 6 Hysteresis observed on Water-Al_2O_3-47nm ($\approx$ 7% particle volume fraction)

We first performed Run 1 (heating phase) that is immediately followed by a cooling phase identified as Run 1a. Viscosity data collected for Run 1 have shown that at $\approx$61.3°C, the nanofluid viscosity reaches its lowest level; beyond this temperature, the viscosity starts to increase with increasing temperature. This behaviour was again confirmed by another run, Run 2 (heating phase). Ones can notice here a quite low dispersion of viscosity data from Runs 1 and 2, which confidently assessed not only the data consistence but also the reliability of instruments and experimental procedures. Then we continued heating the fluid sample beyond 61.3°C and let it slowly cooled afterwards. A striking hysteresis behaviour was then observed. Thus, during the two cooling schemes performed, Run 1a and Run 2a, viscosity values measured for a given temperature are remarkably higher than those collected during the heating phases (Runs 1 and 2). One can then expect that during Runs 1a and 2a, drastic and irreversible damages seem to be induced to the particle suspension properties. It is worth noting that the only difference between Run 1a and Run 2a resides in the maximum temperature reached by fluid sample prior to be cooled: such maximum temperature was approximately 69°C for Run 1a and 71.2°C for Run 2a. One can see from Fig. 6 that values of viscosity corresponding to Run 2a are clearly higher that those of Run 1a. Such result would indicate that the damages to the particle suspension properties are obviously more pronounced in Run 2a. Hence, the temperature level to which fluid sample is heated up has an importance regarding the possible adverse effects on the nanofluid rheological properties. It is worth

mentioning that this intriguing hysteresis behaviour was also observed for other particle fractions as well as for Al_2O_3-water nanofluid with 36nm particle-size. Figure 7 shows in particular the same hysteresis phenomenon observed for CuO-water nanofluid with 29nm particle-size. One can obviously notice that for this particular nanofluid, the increase of viscosity during the cooling phase for fluid sample with 9% particle fraction is simply drastic.
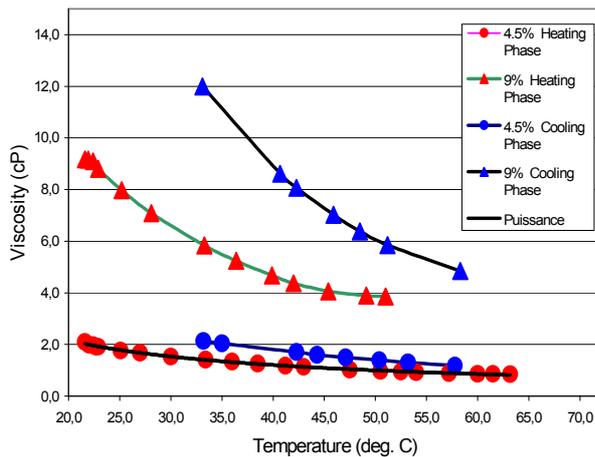
Fig. 7 Hysteresis observed on Water-CuO-29nm (4.5% and 9% particle volume fractions)

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

	Al_2O_3		CuO
	47nm	**36 nm**	**29nm**
1 %	N/A *	N/A *	66.2
4 %	69	65.3	63.2
7 %	63.8	61.6	57.2
9 %	58.7	54	51

Note: N/A* Hysteresis phenomenon not observed for the temperature range considered.

Furthermore, it was observed that when a heated fluid sample is maintained below the critical temperature, then the above hysteresis behaviour did not occur. Table 1 shows the values of the critical temperature as determined experimentally. It has been observed that the critical temperature strongly depends on both the particle volume fraction and particle-size. In fact, T_{cr} has been found to decrease with increasing particle fraction; while for a given particle volume fraction, T_{cr} clearly decreases for smaller particle-size.

The above intriguing hysteresis still remains not very well understood. It is suggested that the considerable increase of viscosity during the cooling schemes 1a and 2a may result from some drastic

and unknown changes that have been caused by temperature effect. In fact, a visual observation of fluid samples at the end of Runs 1a and 2a have clearly shown an evident sign of a highly viscous fluid where the particle agglomeration was quite visible on inner surface of the measuring chamber. This suggests that the particle suspension properties have been greatly altered or, worse, destroyed when the fluid sample is heated beyond the critical temperature. Such a rather interesting fact may closely be linked to the presence of dispersants i.e. chemical agents often used as surfactants to achieve and maintain a good particle suspension. It is believed that beyond a limit temperature, 'expected' effects from these dispersing agents may be greatly reduced or destroyed, which causes particles to lose their suspension capabilities. Particles have then tendency to agglomerate that, in turn, may result in an erratic and drastic increase of the nanofluid viscosity as observed. Such an explanation appears, to our opinion, quite plausible as it can explains that once the deterioration of the particle suspension properties is initiated, the presence of more particles within the base fluid - i.e. in case of higher particle volume fractions (for given particle-size) or smaller particle-size (for given particle volume fraction) - would logically result in a more pronounced effect on the nanofluid viscosity. This seems to be well corroborated through the values of T_{cr} shown in Table 1. Unfortunately, it was not possible to provide any information regarding the nature and characteristics of the dispersants used by the manufacturer. More investigations will be indeed needed in order to completely understand such striking phenomenon. Finally, the viscosity database presented, which is believed among the first of a kind, constitutes, to our opinion, an important contribution. The hysteresis behaviour on nanofluids, which has not yet been reported in the literature, can raise serious concerns regarding the use and limitation as well as the manufacturing process of today nanofluids for the heat transfer enhancement purposes.

4   Conclusion

A new and complete viscosity database has been established for three particular water-based nanofluids, namely 36nm and 47nm-Al_2O_3-water and 29nm-CuO-water. Data have shown that in general, the nanofluid viscosity strongly depends on both temperature and particle volume fraction; while the particle-size effect seems to be important only for high particle fractions. The existence of the

critical temperatures has been experimentally established. When heating beyond such critical temperatures, the particle suspension properties seem to be drastically altered, which in turn, has triggered the hysteresis behaviour on nanofluids. Such a striking phenomenon raises serious concerns regarding the practical use of the actual nanofluids for the heat transfer enhancement purposes.

Acknowledgements

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