Possible explanations of the deviations in the thermal conductivity measurements

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ABSTRACT

Ever since the report of the abnormal thermal conductivity enhancement of nanofluids by the late S.U.S. Choi, many researchers have observed the extraordinarily high thermal conductivity. However, there are deviations between research groups in the effective thermal conductivity measurements, which leaded researchers to doubt about the using nanofluids as an alternative heat transfer fluid.

In this study, the cautions in thermal conductivity measurements using transient hot-wire method, and the non-equilibrium nature of nanofluids were elucidated as possible explanations of the deviations. It was found to be very important to avoid the impacts of finite wire thermal capacity and natural convection during the thermal conductivity measurement using transient hot-wire method, and the particle contents as well as its size distribution in base fluids should be always monitored and reported as the measurement results of nanofluid thermal conductivities.

Keywords: Nanofluids, thermal conductivity, transient hot-wire method

1 INTRODUCTION

Nanofluids were invented to enhance thermal conductivity of the working fluid near the wall region, where the conduction is the only mechanism to dissipate heat and it limits the heat transfer rate from a body to fluid. Many research groups have reported thermal conductivity measurements of homemade nanofluids, although there were large deviations between them, without investigating the causes. This presentation explains the apparent deviations between measurement results in terms of the fundamental non-equilibrium nature of nanofluids.

Recently, Hong *et al.* [1] and Han *et al.* [2] performed critical studies on the transient hot-wire method, which has been commonly used by many research groups due to its simplicity and short response time. They reported that the proper selection of the temperature history range is crucial to avoid the impact of natural convection on the thermal conductivity estimation, and the temperature dependence of nanofluid thermal conductivity enhancement observed by other research groups might be misleading due to the failure to prevent it.

Thermal conductivity of a pure substance is a physical property which is determined under a thermodynamic equilibrium state. However, even the ideal nanofluids are in a metastable state. A consequence of this is that it takes a very long time to reach their equilibrium conditions, the segregated state between the particles and base fluid. Real nanofluids suffer from sedimentation and aggregation resulting in the variation of particle volume fraction and particle size in time. Han [3] investigated the effects of sedimentation and aggregation on the nanofluid thermal conductivity. The changes of particle volume fraction and its size were monitored using hydrometers and dynamic light scattering method respectively. He reported that the particle volume affected the thermal conductivity more for the nanofluids studied, and introduced a way to distinguish the impact of the particle volume and its size.

The possible explanation of the deviations in thermal conductivity measurements of different research groups will be presented, which is a proposed standard to measure the nanofluid thermal conductivity and a way to describe the thermodynamic state of nanofluids.

2 PROBLEMS IN MEASUREMENTS

To measure the thermal conductivity of fluids using transient hot-wire method, electric power is introduced to the hot-wire where it is converted to heat which increases the hot-wire temperature. The amount of heat and the thermal conductivity of the surrounding fluid determine the temperature changes in the wire. For fluids of high thermal conductivities, the heat dissipates well and hence results only in a small hot-wire temperature rise. In contrast, higher hot-wire temperatures can be expected when the surrounding fluid's thermal conductivity is small. The thermal conductivity of the fluid is measured under the assumption that heat transfer occurs only in the conduction mode. The relation between the hot-wire temperature history and the thermal conductivity of the fluid is given in Hong *et al.*[1] and Han *et al.*[2] as:

$$k_f = \frac{\dot{Q}}{4\pi L_w} \cdot \frac{d\ln t}{dT_w} \tag{1}$$

2.1 Finite Wire Thermal Capacity

Figure 1 shows the effect of actual heat conduction inside the wire. The values of thermal conductivity were obtained from the measured hot-wire temperature history data using Eq. (1) in which the heat flux on the surface is set to be a constant. However, it takes a finite time for the heat, generated inside the wire, to reach the wire surface uniformly. The 'ideal' heat flux history, which neglects the time delay for the internally generated heat to arrive at the surface, and 'real' heat flux history which is obtained from the simulation results, are compared in Fig. 1. It takes around 1 s for the surface heat flux to reach steady state; thus, the thermal conductivity will be overestimated if the wire temperature history data before 1 s are used. In our case, platinum hot-wires of 50 nm in diameter coated by Teflon of 25 nm in thickness were used. Clearly, for thicker wires it will take longer for the surface heat flux to arrive at the steady value. As a result, the temperature history data affected by the surface heat flux time delay should be avoided when using THWM; otherwise, measurements would result in overestimation of the thermal conductivity.

2.2 Avoidance of Natural Convection Impact

As the hot-wire temperature increases due to the Joule heating effect, the temperature difference between the hot-wire surface and the bulk fluid increases. The temperature difference generates a buoyancy force due to the density difference between the fluid on the hot-wire surface and the bulk fluid, which is balanced by the viscous force in the early stage. With further increase of wire temperature, the buoyance force exceeds the viscous force and natural convection starts to occur in the test cell. As in addition to heat conduction also natural convection comes into play in the course of thermal conductivity measurements using THWM, the hot-wire generating heat will be removed to the surrounding fluid more rapidly. The increased heat transfer rate will result in a smaller hot-wire temperature
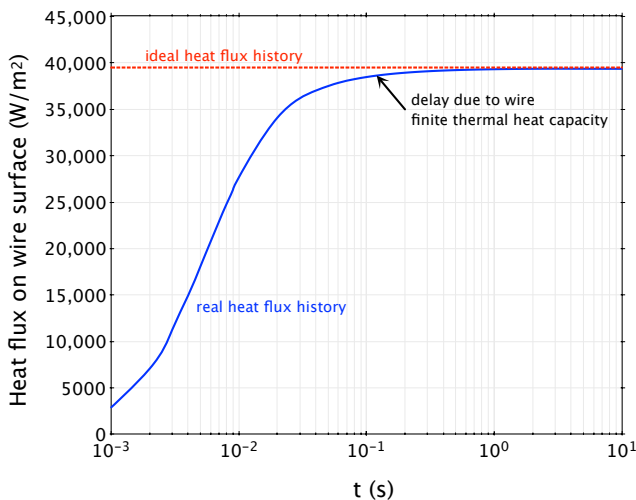
rise over time, which will lead to an increase in the gradient term in Eq. (1), resulting in an overestimation of the thermal conductivity. Therefore, it is necessary to restrict the end time of the temperature history used for data processing to the point where measurements are not affected by natural convection. In order to determine the unaffected region, thermal conductivities are measured by varying the start- and end-times. The slopes in Fig. 1 decrease when taking the temperature history data at a later part which is affected by natural convection. As the start time delays, the impact of natural convection on the thermal conductivity becomes stronger and the slope becomes smaller for a fixed end time. To identify the point where the natural convection effect comes into play, the following procedure has been used. First, thermal conductivities are obtained for different end-times, keeping the start-time fixed. Then, the thermal conductivities are estimated for varying start-times. With larger start-times the slope of the increasing thermal conductivity increases as well, generating "cross-over points" beyond which the effect of natural convection cannot be ignored. Thus, any temperature data beyond the cross-over points should be effectively eliminated (see Hong *et al.* [1]).

Han *et al.* [2] and Han [3] claimed that the temperature dependence of nanofluid thermal conductivity enhancement was found to be caused by the failure to remove the natural convection impact on the measurements. Avoiding the effect, the thermal conductivity enhancement was found to be independent of fluid temperature.

In summary, special care should be taken of in measuring the nanofluid effective thermal conductivity; (a) the temperature should be taken after the initial heat flux delay period, (b) the end time of the temperature history should not go beyond the "cross-over" point not to be affected by natural convection, and (c) the temperature history length should be long enough not to be affected by the noise caused by either the differentiation operation in Eq. (1) or the power source. Please refer to Hong *et al.* [1] and Han *et al.* [2] for more details.
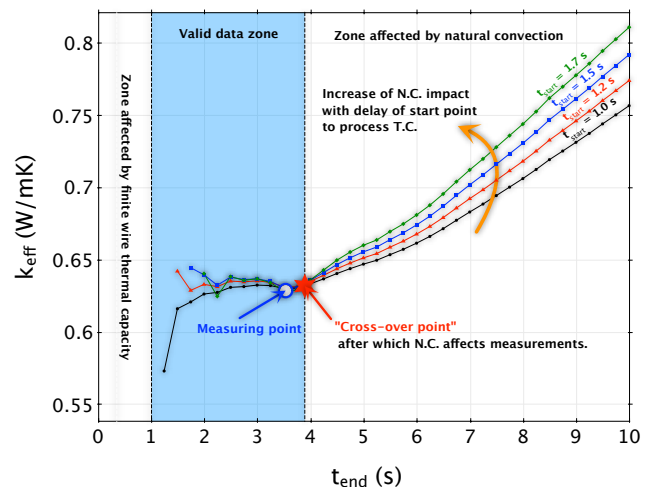

Figure 1: Heat flux history on the surface of the hot-wire

Figure 2: Valid temperature history range to estimate

thermal conductivity free from natural convection effect.

3 NON-EQUILIBRIUM NATURE OF NANOFLUIDS

3.1 Particle Sedimentation in Time

Water-base Ag and EG-base ZnO nanofluids were prepared by the one-step method. They were re-dispersed by applying ultra-sonication for three hours using a bar-type sonicator. Since the suspended particle volume fraction could vary in time, the particle volume fractions were monitored by measuring the specific gravity of the nanofluids, using an hydrometer (GP-300S, Matsuhaku, Japan). Then, the volume fractions were obtained by the following equation:

$$\phi = \frac{\rho_n - \rho_f}{\rho_p - \rho_f} \qquad (2)$$

where ϕ, ρ, n, p and f represent particle volume fraction, density, nanofluid, particle and base fluid. With the particle and liquid densities known, that of each nanofluid could be estimated from the specific gravity measured by the hydrometer.

Figure 3 represents the particle concentration, or volume fraction variation in nanofluids in time. It shows the contrast of the stabilities of EG-base ZnO and water-base Ag nanofluids. The volume fraction of EG-base ZnO nanofluid decreased drastically for the first week due to sedimentation, while that of water-base Ag nanofluid stayed at a constant value. Usually, researchers estimate the volume fraction at the time of manufacturing nanofluids from the information of particle mass added into basefluid and particle & fluid densities, which have been reported in the articles so far. It is for sure that the volume fraction of nanofluids decreases with time, so that it is very important to report the volume fraction of the nanofluids at the time to be tested for the thermal conductivity.
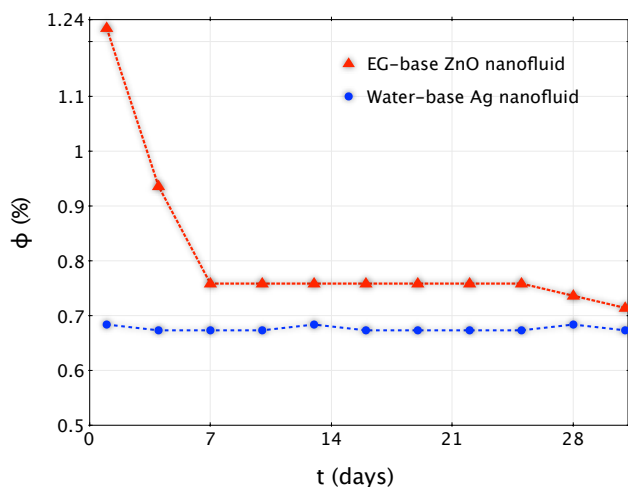

Figure 3: The effect of sedimentation on particle volume fraction (ϕ) variations of two nanofluids in time

3.2 Particle Growth in Time

Many researchers, e.g., have reported that the nanofluid conductivity is a function of particle concentration, particle size and others. Therefore it is important to report the particle size as well as the volume fraction when researchers report the results of their measurements.

Figure 4 shows the change of particle size in time. The number weighted average particle size was analyzed multiple times by dynamic light scattering method, and the results are shown in the figure. It was observed that EG-base ZnO nanofluid suffered from the aggregation to result in particle size increase, whereas the particle size of water-base Ag nanofluid remained small and constant. The effect of aggregation may couple with that of sedimentation, since the larger particles tend to be affected more by the impact of the gravity.
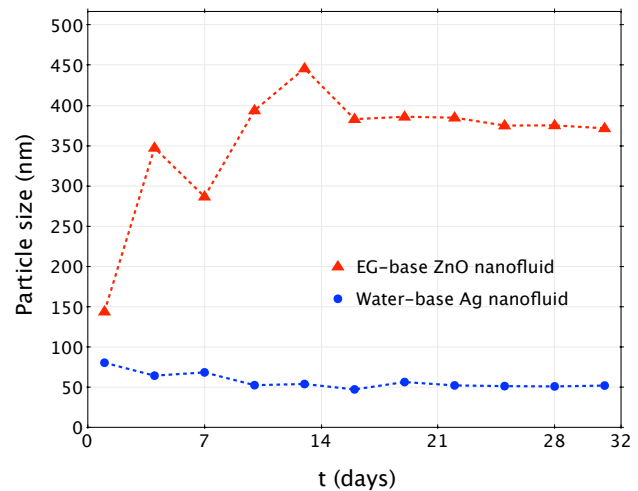

Figure 4: The effect of aggregation on particle size in time

3.3 Thermal Conductivity Change in Time

Figure 5 represents the thermal conductivity variations in time. The thermal conductivity enhancement of EG-base ZnO nanofluid decreased sharply during the first ten days and decreases gradually after that, which could be explained by the change of the volume fraction and particle size in time. The thermal conductivity enhancement of water-base Ag nanofluid remained at a constant value, as expected from the behaviors of volume fraction and particle size in time. Nanofluid thermal conductivities were still greater than those estimated by the conventional Maxwell model. It was noticed that the thermal conductivity enhancement of water-base Ag nanofluid estimated by Maxwell model was lower than that of EG-base ZnO nanofluid, which the other way was true in measurement results. This might be the unique characteristics of nanofluids, which could be attributed to the smaller
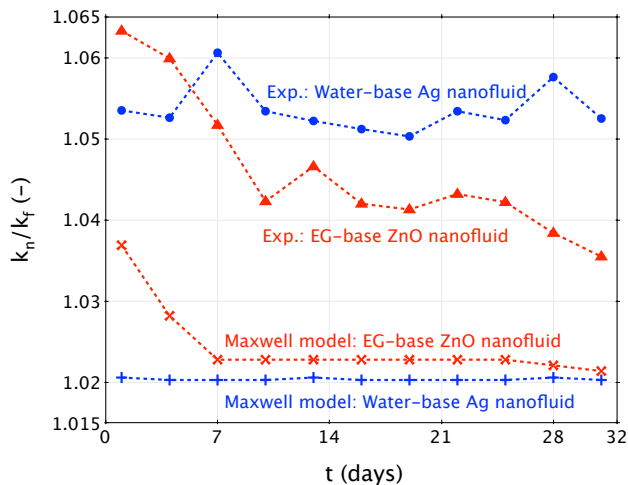
Figure 5: The effect of aggregation on particle size in time

hydrodynamic particle size of Ag nanoparticles in water, where the nanoscience and nanoengineering could get envolved to enhance the performance.

There is another point to be stressed. The thermo-dynamically stable state of particle suspensions is the segregation into the particle and fluid phases. Although the time to reach the segregated state could differ from one to another, all nanofluids will end up with the separation of phases. Therefore, it might not be a good idea to measure the thermal conductivity of nanofluids. Thermal conductivity is a thermodynamic property of matter, which is defined at the thermodynamic equilibrium conditions. The concept of the effective thermal conductivity started from the attempt to model the nonofluid thermal conductivity as that of a single phase material due to the dilute or negligible amount of particles in fluids. In future, the validity of using the effective thermal conductivity concept should be tested rigorously by researchers.

4   CONCLUSIONS

In this work, two possible explanations of deviations in nanofluid thermal conductivity measurements were presented.

- The mistakes in the selection of proper temperature history in the measurements using transient hot-wire method were discussed as the first possible explanation of deviations in measurements between researchers. To avoid this, special care should be taken of in measuring the nanofluid effective thermal conductivity; (a) the temperature should be taken after the initial heat flux delay period, (b) the end time of the temperature history should not go beyond the "cross-over" point not

to be affected by natural convection, and (c) the temperature history length should be long enough.

- The non-equilibrium nature of nanofluids was the other possible cause of the deviations. It was recommended that the researchers provide the particle volume fraction and the particle size when they report the results of the measurement in the future.

- The thermal conductivity is a concept defined in thermodynamic equilibrium condition, and it might not suit for the cases of nanofluids, which are in non-equilibrium state in nature. Further rigorous research should be done to testify the validity of using it.

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