Nanofluids – potentials and illusions

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Abstract

This study is motivated by the controversies on nanofluids reflected in recent literature. Eight hypotheses considering the subject's measurement of thermo-physical properties, evaluation of heat transfer experiments and numerical modeling are discussed. Two of them are illustrated employing the most recent experimental results by the authors.

Keywords: nanofluids, experimental difficulties, further work.

1 Introduction

Confronted with limited energy and material recourses and undesirable manmade climate changes science is searching for new and innovative strategies to save, transfer and store thermal energy. One of the currently most intensively discussed options are the so-called nanofluids. The number of publications with respect to nanofluids is exponentially growing since the beginning of our millennium (Fig. 1). However, many publications indicate controversial results.

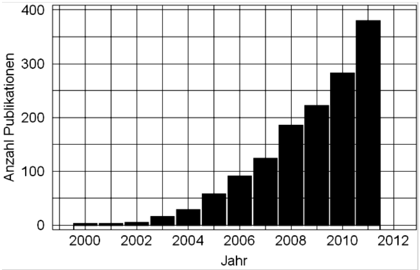

Figure 1: Publications with respect to the key word nanofluids according to ISI Web of Knowledge (data taken on January 12th, 2012).

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A general lack of theoretical understanding describing the experimentally found results is obvious and retards successful technical applications. This study aims to compile some of the possible reasons causing these controversies.

Nanofluids are suspensions consisting of a liquid basefluid and solid particles. The particles have a size ranging from 10 to 200 nm. Such nanoparticles are much larger than water molecules which have a size of about 0.1 nm. However, they are also much smaller than particles in the micrometer and millimeter range. The diminutiveness of the nanoparticles gives hope that they do not disturb applications by clogging, sedimentation and abrasion. The particle materials employed among others are pure metals, ceramics (Al_2O_3, SiO_2, etc.) and carbon (nanotubes, soot, and diamante). The general expectation is that the higher thermal conductivity of these materials leads to an effectively increased thermal conductivity which in turn should enhances heat transfer.

Currently the number of publications with respect to nanofluids *per annum* counts to several hundredths which makes it nearly impossible to extract major trends from this tremendous body of literature. Therefore, Sergis and Hardalupas [1] employed statistical tools to analyze 130 recent publications. Their major finding is that nanofluids behavior cannot be described well with customarily physical models so far. However, this should not mislead about the fact that the majority of experiments indeed indicate increased thermal conductivity and enhanced heat transfer. Sergis and Hardalupas [1] find that about 75% of the analyzed studies point out an increase of thermal conductivity of the investigated nanofluids between 1 and 24% (30% between 5 and 9%) compared with the basefluid. As regards to convective heat transfer about 18% of the authors prove an enhancement between 10 and 19%. Another 45% of the studies describe a quantitatively not specified enhancement. Only about 11% of the authors are hostile to an enhancement of heat transfer. For phase changes namely pool boiling about 50% of the studies see an improvement. These numbers clearly underline the chances nanofluids offer. However, there are also a comparable large number of negative results and critical papers (e. g. Putnam *et al.* [2], Walsh *et al.* [3], Sommers and Yerkes [4]) which do rather see nanofluids as an illusion at least for turbulent flow (Prabhat *et al.* [5]).

2 Why produce nanofluid experiments so different results?

In the following eight hypotheses will be discussed which shall help to explain why experimental and numerical investigations of nanofluids indicate so different results. Some of these hypotheses are indeed facts. Of course the hypothesis provided are incomplete and based on the limited experiences of the authors. However, they are neither assumptions nor speculations. They rather follow directly from experiments carried out at ILK, scientific disputations with respectable colleagues or careful analysis of a large number of recent publications.

2.1 Thermophysical properties

2.1.1 Nanofluids are complex two-phase liquids

Nanofluids are indeed very complex two-phase composites consisting of basefluid and nanoparticles. The effective thermophysical properties are constituted by both the thermophysical properties of basefluid and particles. An exception is the dynamical viscosity because nanoparticles are solid and their properties do not contribute directly to this parameter. However, the particles themselves change the viscosity just by being part of the nanofluid. Beside the nanoparticle concentration, the particle morphology (size, shape, fractal surface etc.) and their behavior under imposed outer gradients (e.g. thermophoresis) and forces (e.g. magnetic forces Shima and Philip [6]) are additional factors which influence the properties. Therefore different thermophysical properties might be caused by small differences of the compared nanofluids which are either not considered or which are simply declared negligible. It is a matter of fact that the possible mechanism for increased thermal conductivity (Keblinsky *et al.* [7]) and enhanced heat transfer are manifold and theoretically still not well understood.

2.1.2 Chemical ingredients change base fluid properties

The third component beside basefluid and nanoparticles influencing the thermophysical properties of nanofluids is the sum of all chemical ingredients (dispersant agents, surfactants etc.) employed to stabilize the suspension. To be fair not all producers of nanofluids employ such substances. For example the gold nanofluids by Kim *et al.* [8] obtained by a one-step laser ablation technique show an outstanding colloidal stability although no dispersants were employed. However, in case such substances are added the properties of the basefluid are changed. If these changes are weak or strong might be different in each individual case. Nevertheless the direct comparison of the effective thermophysical properties of the nanofluid with the equivalent properties of the original basefluid seems to be questionable. Seemingly increases or changes might be simply caused by the changed basefluid properties or better the properties of the liquid matrix filling the space between the nanoparticles.

Unfortunately the comparison of effective thermophysical properties with the equivalent properties is common practice. It goes back to the classical models for the dynamical viscosity and the thermal conductivity by Maxwell (1873) and its extension by Nan *et al.* [9]. A discussion of more recent viscosity models can be found in Kole and Dey [10] and for thermal conductivity in Ding *et al.* [11].

2.1.3 Particle size and agglomeration effects are underestimated

It is difficult to understand how different production technologies namely one-step procedure (*in situ* generation of nanoparticles) and two-step procedure (nanoparticles are produced separately and later dispersed in the basefluid) can create different nanofluid properties when the same ingredients are used. Assuming that indeed no or the same chemical substances are added to stabilize the nanofluid the only explanation would be particle agglomeration. Agglomeration of primary particles produces not only larger particles but generates also new shapes, different aspect ratios and surface topologies.

Timofeeva *et al.* [12] investigated two-step nanofluids with respect to the influence of the particle shape on viscosity and thermal conductivity. They found that the enhancement of the thermal conductivity forecasted by several theoretical approaches is significantly diminished due to interfacial effects proportional to the surface area of nanoparticles. However, they also showed that viscosity and thermal conductivity indeed depend on the shape of the primary particles. Viscosity is for example strongly increased by agglomerates with large aspect ratio. In general one can conclude that especially for two-step procedures the size and shape of the primary nanoparticles are different from size and shape of the nanoparticles indeed acting within the nanofluid.

2.1.4 Nanofluids behave as such even in measurement devices

Nanofluids are two-phase suspensions. The common hopes respectively believe are that due to the minuteness of the particles nanofluids behave like single-phase liquids. However, when determining the thermal properties of a nanofluid experimentally the two-phase character cannot be excluded *a priori*. It has rather to be proven that the assumption of a single-phase liquid is reasonably. Effects related to the two-phase character (liquid/solid) namely sedimentation, thermophoresis, enhanced Brownian motion and hydrodynamic diffusion may affect measurements of dynamical viscosity and thermal conductivity.

Sedimentation follows from the imbalance of forces acting on nanoparticle. It is described by Stoke's law which summarizes the constant weight force, the buoyancy force and the velocity dependent friction force. In case these forces are balanced the nanoparticle would float if not nanoparticles sink and eventually build sediment on the bottom of the measurement device.

Thermophoresis is caused by temperature gradients occurring when measuring temperature dependencies of thermophysical properties. Of course a constant temperature within the measurement volume is aimed. However, to achieve the thermally balanced state takes some time and nanoparticles are subjected to spatial temperature gradients. More intensive Brownian motion on the hot side of the particles pushes them against the direction of the temperature gradients. Similarly enhanced Brownian motion occurs for example in classical rotary viscometers when heated to measure viscosity at higher than ambient temperature. Nanoparticles may then be driven together and the rate of agglomeration is increased. Under such circumstances nanofluids change their properties during the measurement campaign (Sec. 3.1). Hydrodynamic diffusion occurs due to hydrodynamic interaction (Buggisch and Muckenfuss [13]). The nanoparticles drift across the shear direction and build consequently a concentration profile which in turn causes varying properties.

Measurement devices and research teams have to cope with the physical effects mentioned above and others. For illustration the commonly employed device for the measurement of the thermal conductivity the KD2 Pro (Decagon Devices, http://issuu.com/decaweb) is discussed. This apparatus is primarily designed for measuring thermal conductivity and thermal resistivity of soil, concrete and rock but allows also the measurement of the thermal conductivity of liquids if the KS-1 sensor is used. The measured thermal conductivity should than be smaller than 0.1 W/m-K (p. 7, Operator's manual). At ambient temperature water has a thermal conductivity of about 0.6 W/m-K. Further the manual states that "error from convection heat exchange is often very large, rendering the thermal properties measurements useless, and must be avoided" (p. 46). Paul *et al.* [14] employed the KD2 Pro to measure thermal conductivity of water based gold nanofluids. They found an increase of the thermal conductivity depending on the particle size up to 45%. The measurement were repeated by a team from IBM (Shalkevich *et al.* [15]) employing a static heated plate and a transient hot wire method and in our own laboratory employing a ring gap apparatus (for description see Ehle *et al.* [16]). The maximum increase of thermal conductivity was found to be about 1.4%. Note that this result follows from two independent institutes employing three independent measurement techniques. To summarize the KD2 Pro is of course an excellent tool for measuring construction and insulation material but seems to be not made for academic research.

2.2 Heat transfer

2.2.1 Erroneous scaling leads to wrong conclusions

Physical experiments are carried out to answer questions. With respect to nanofluids two questions are most important.

i. Is heat transfer of nanofluid flow enhanced compared to the basefluid flow?
ii. Is the enhancement if there is one *anomalous*? Or in other words do unexpected physical mechanisms namely two-phase flow effects affect heat transfer?

The first question is mostly easily to answer. Experiments have to be done twice once with the pure basefluid and second with the nanofluid. Than dimensional parameters of interest like outlet temperature [K], amount of transferred heat [W] or pressure loss [Pa] have to be compared at identical pumping powers [W] or mass fluxes [kg/s]. The *better* fluid is simply the one which has the higher outlet temperature, the higher amount of heat transferred etc.

The second issue is rather difficult because it is basically the question for the essential physical mechanisms. If these mechanisms cannot be identified directly from the experiments additional tools like similarity analysis based on Buckingham's Π-theorem (Gersten and Herwig [17]) have to be employed. The standard example for this is the thermally developing pipe flow in a straight circular pipe. The semi-analytical solution provided by Nusselt [18] is the exact solution of the partial differential equations governing the problem.

The heat transfer of any ordinary single-phase Newtonian fluid should be described in more or less agreement with [18]. In case nanofluid flows indicate higher Nusselt numbers than predicted additional physical effects not gathered by the governing equations must be considered. Of course such a result can only be obtained when flow situations having identical Graetz numbers x^+ are compared. Comparing flow situations having merely identical Reynolds or Peclet numbers would be misleading. In both cases complete fluid mechanical and thermodynamical similarity would not be given.

Unfortunately most flow situations are much more complex than the thermally developing laminar entrance flow. Mostly it is not known uniquely which similarity numbers or combinations of them characterize full similarity sufficiently. Here the example *par excellence* is the laminar pipe flow with inserted twisted tape. Manglik and Bergles [20] compiled a nice collection of Nusselt number correlations describing this special type of flow. These correlations mostly contain Reynolds and Prandtl number as products of the form $Re^m Pr^n$. According to this the Nusselt number would increase by merely increasing the Prandtl number and keeping Reynolds number and geometry unchanged. Exactly that happens with nanofluids. The Prandtl number of these liquids is higher than the one of water. The reason is mainly that dynamical viscosity is more increased than thermal conductivity.

Therefore a comparison merely based on identical Reynolds numbers would be misleading. Either experiments having identical Reynolds numbers as well as identical Prandtl number or experiments having identical $Re^m Pr^n$-values must be compared. Regrettably this strategy is not always accepted and comparisons based on Reynolds number alone are employed customarily (e. g. Pathipakka and Sivashanmugam [21], Wongcharee and Eiamsa-ard [22]).

2.2.2 The dominating heat transfer mechanism is essential

The Nusselt number of laminar flow through channels with different cross sections has mostly an order of magnitude of about $O(1)$ (Shah and London [X]). One of the oldest and simplest correlations describing the Nusselt numbers of turbulent pipe flows is the Colburn analogy (Jiji [19]). Considering pure water with a Prandtl number of about 7 at ambient temperature turbulent Nusselt numbers reach values of an order of magnitude of $O(10)$ ($Re_D = 10^4$) and of $O(100)$ ($Re_D = 10^5$). This simple exercise shows that turbulent heat transfer is at least by a factor of 10 to 100 larger than its pure laminar counterpart. The reason therefore is clear. While in pure laminar flow conduction is dominant turbulent exchange processes close to the wall enhance turbulent heat transfer significantly. The same is true for any classical turbulence generator. The dominant turbulent heat transfer mechanism is the wall normal transport of. Such transport is much more effective than conduction.

Based on the aforementioned one can ask if a nanofluid just due to its increased thermal conductivity say by a factor of 1.01 to 1.2 or so could indeed enhance turbulent heat transfer. Or does it rather suppress turbulent exchange processes close to the wall due to its increased viscosity? From this simple argumentation one can conclude if an increase in turbulent heat transfer of nanofluids is observed it should be due to the two-phase character of the flow. However, a most recent critical survey of twelve pipe flow experiments with nanofluids (eight laminar and four turbulent) by [5] indicated that an increase of Nusselt number might occur only in laminar flow especially in the entrance region.

2.3 Numerical modeling

2.3.1 Bottom-up-models superior to top-down models

The two-phase character of nanofluid flows cannot be disregarded *a priori* when it comes to numerical models. Any numerical model should start with a set of equations completely describing a two-phase flow situation. Additional terms and/or equations for Brownian motion, thermophoresis etc. have to be considered explicitly. In such a bottom-up-approach nanoparticle concentration becomes a field property. Avramenko *et al.* [24] showed with their Lie-group based numerical model for laminar boundary layers the effects of local nanoparticle concentrations on velocity and temperature profiles, as well as on Nusselt number. Similarly Heyhat and Kowsary [25] proposed based on a two-phase numerical model that the enhancement of heat transfer in laminar pipe flow is not only due to the increase of the effective thermal conductivity but also due to nanoparticle migration.

The alternatives are top-down-approaches which assume right from the beginning that nanofluids behave as single phase liquids. Only the correlations for the properties are changed according to some model assumptions or experimental findings. Unlike bottom-up-models the results of such a model do not offer the chance to validate the single-phase-flow-assumption. Only when bottom-up-models predict that the two-phase flow effects are weak and negligibly this is assumption is reasonable.

2.3.2 Thermophysical property correlations must follow experiments

For the properties density, specific heat capacity and thermal expansion coefficient simple weighting functions exist which allow the calculation of the effective nanofluid properties (e.g. Ben Mansour *et al.* [26]). A large number of correlations exists for dynamical viscosity (e.g. Ding *et al.* [11]) and thermal conductivity (e. g. Kole and Dey [10]). However, no general model seems to be found so far. The reason is mainly the diversity of nanofluids (Sec. 2.1.1 and 2.1.2) and a limited knowledge with respect to the mechanisms constituting these properties. However, results of numerical simulations depend definitively on the quality of the correlations describing thermophysical properties. Sensitivity studies carried out by He *et al.* [27] and Abu-Nada [28] for different ceramic/DI-water nanofluids confirm this. The consequence is that currently only property correlations which are based on experimental results should be implemented in numerical codes.

3 Examples

In the following two examples are presented which illustrate the hypothesis discussed in Sec. 2.1.4 and Sec. 2.2.1. Both examples are based on experimental studies carried out at ILK Dresden.

3.1 Thermal conductivity of a gold nanofluid

Thermal conductivity of two DI-water based gold nanofluids was measured employing the ring gap apparatus described in detail by Ehle *et al.* [16]. The nanofluids were produced by Particular GmbH (Germany) employing a one-step laser ablations technology. Both suspensions had a nanoparticle concentration of 5×10^{-4} vol. % and were mildly stabilized with 0.07 wt% sodium citrate. Mean values of nanoparticle sizes were 5 nm respective 60 nm (DLS-measurements). The color of the fresh samples was ruby. Optical inspection with naked eye after measuring viscosity respectively thermal conductivity showed that the color of both nanofluids had altered to different types of brownish red. The color changes were caused by changed light refraction due to increased particle sizes.

Figure 2 (left) compares the experimentally found increase of thermal conductivity in dependency on temperature with results by Buongiorno *et al.* [29] and Shalkevich *et al.* [15]. Despite the small number of available data a reasonable agreement is found. Surprisingly deterioration is observed with increasing temperature. However, it seems to be not the increasing temperature which causes this deterioration. The right plot of Fig. 2 shows the increase of the thermal conductivity chronologically ordered according to the experimental sequence. While the 5 nm gold nanofluid was measured every 5 Kelvin (20°C, 25°C ... 60°C) the temperature sequence of the 60 nm sample was 20°C, 30°C, 40°C, 50°C, 60°C, 55°C, 45°C, 35°C and 25°C. In both cases the first three measurements show an increase of about 1%. Later measurements scatter around the zero line. Therefore it seems to be more likely that the time the samples were

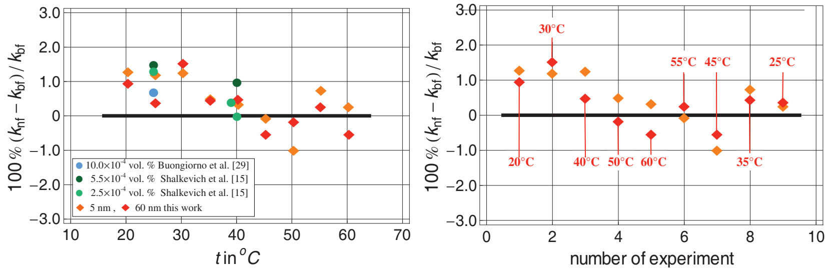

Figure 2: Increase of thermal conductivity in comparison with other experiments (left) and enhancement of thermal conductivity according to the experimental chronology. In the right plot the temperature of the measurements of the 5 nm gold suspension (orange symbols) increases by 5 K from left to right.

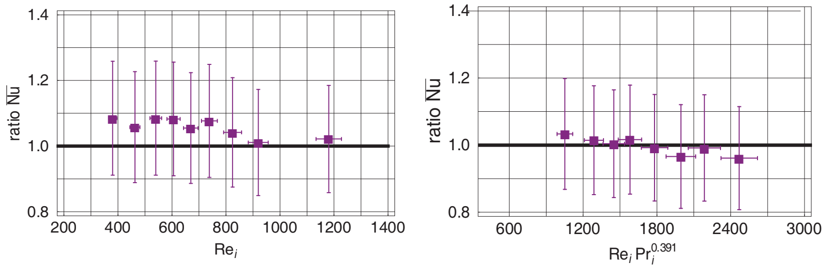

Figure 3: Ratio of averaged Nusselt number of nanofluid to pure water for pipe flow with inserted twisted tape. Scaling based on Reynolds number alone (left) and scaling based on Reynolds and Prandtl number (right).

exposed to heating changes their morphology. In agreement with Sec. 2.1.4 it is hypothesize that enhanced Brownian motion leads to irreversible particle agglomeration which would also explain the changed colors.

3.2 Scaling of laminar twisted tape flow

Nanofluids are not only interesting due their increased thermal conductivity but also due to the possible enhancement of heat transfer. Of special interest is the question if enhancement techniques as twisted tapes could be even more effective when combined with nanofluids? In a recently finished research project such twisted tapes were investigated by ILK. The twisted tape was made of a 1 mm thick copper sheet and had a dimensionless twist ratio of about 6. Beside DI-water a TiO_2 nanofluid with a nanoparticle concentration of 5 vol. % (d_{np} = 30-80 nm) was investigated. Figure 4 shows the ratio of the averaged Nusselt number of the TiO_2 nanofluid to DI-water flow.

The left diagram of Fig. 3 compares flow cases having nearly identical Reynolds numbers (±10%). From that plot one could conclude that the nanofluid Nusselt number is larger than the DI-water Nusselt number due to *anomalous* (namely two-phase flow) effects. A Nusselt number ratio of unity would indicate that the nanofluid heat transfer coefficient is increased in the same amount as the thermal conductivity. The increase of the thermal conductivity of the TiO_2 nanofluid is about 11%. A further conclusion from the plot under discussion would be that the Nusselt number is increased by about 8% for Re_i less than about 700. Above this threshold a deterioration of the effect seems to occur.

Allowing a scaling based on Reynolds and Prandtl number of the form $Re^m Pr^n$ as proposed by empirical correlations delivers a completely different picture (Fig. 3 right). For m = 1 and n = 0.391 the ratio of the Nusselt numbers is now nearly unity throughout the entire range investigated. Note that now different pairs of flow realizations than for the Reynolds number based scaling are compared. From this plot it is concluded that there is no Nusselt number increase and the heat transfer coefficient is increased in the same amount as the thermal conductivity. There are no additionally two-phase flow effects acting. It is rather an effect following from the changed thermophysical properties which is entirely described by the Prandtl number.

4 Conclusion

It is not unusual that experimental, theoretical and numerical results from different scientific studies vary. This is especially true when the subject of interest is complex and comparably new. Even *ordinary* research topics like the turbulent flow of a Newtonian fluid in a straight pipe which is intensively investigated for more than a century and employed in myriads of technical applications suffers from controversial debates [30]. Therefore none of the hypotheses discussed above should be called negative, dramatic or even the end of nanofluids. It is rather the usual way science has to go when it comes to new and complicated subjects. Science is indeed a sort of a maze and we have to start over and over again to find out what are the real physics behind our research subjects. However, experimental and numerical work should take into account the state-of-the-art knowledge. Nanofluids are so complex that it seems to be necessary that they have to be tailor made to be successful. Experiments and simulations should be carried out which indeed allow investigating effects occurring on length and time scales related to the nanoparticle.

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