Critical Evaluation of Anomalous Thermal Conductivity and Convective Heat Transfer Enhancement in Nanofluids

By

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Indian Institute of Technology Bombay

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By

Naveen Prabhat

Submitted to the Department of Nuclear Science and Engineering on May 13, 2010 in Partial Fulfillment of the requirements for the Degree of Master of Science in Nuclear Science and Engineering

ABSTRACT

While robust progress has been made towards the practical use of nanofluids, uncertainties remain concerning the fundamental effects of nanoparticles on key thermo-physical properties. Nanofluids have higher thermal conductivity and single-phase heat transfer coefficients than their base fluids. The possibility of very large thermal conductivity enhancement in nanofluids and the associated physical mechanisms are a hotly debated topic, in part because the thermal conductivity database is sparse and inconsistent. This thesis reports on the International Nanofluid Property Benchmark Exercise (INPBE) in which the thermal conductivity of identical samples of colloidally stable dispersions of nanoparticles, or ‘nanofluids’, was measured by over 30 organizations worldwide, using a variety of experimental approaches, including the transient hot wire method, steady-state methods and optical methods. The nanofluids tested were comprised of aqueous and non-aqueous basefluids, metal and metal oxide particles, near-spherical and elongated particles, at low and high particle concentrations. The data analysis reveals that the data from most organizations lie within a relatively narrow band (±10% or less) about the sample average, with only few outliers. The thermal conductivity of the nanofluids was found to increase with particle concentration and aspect ratio, as expected from classical theory. The effective medium theory developed for dispersed particles by Maxwell in 1881, and recently generalized by Nan et al., was found to be in good agreement with the experimental data.

The nanofluid literature contains many claims of anomalous convective heat transfer enhancement in both turbulent and laminar flow. To put such claims to the test, we have performed a critical detailed analysis of the database reported in 12 nanofluid papers (8 on laminar flow and 4 on turbulent flow). The methodology accounted for both modeling and experimental uncertainties in the following way. The heat transfer coefficient for any given data set was calculated according to the established correlations (Dittus-Boelter’s for turbulent flow and Shah’s for laminar flow). The uncertainty in the correlation input parameters (i.e. nanofluid thermo-physical properties and flow rate) was propagated to get the uncertainty on the predicted heat transfer coefficient. The predicted and measured heat transfer coefficient values were then compared to each other. If they differed by more than their respective uncertainties, we called the deviation anomalous. According to this methodology, it was found that in nanofluid laminar flow in fact there seems to be anomalous heat transfer enhancement in the entrance region, while the data are in agreement (within uncertainties) with the Shah’s correlation in the fully developed region. On the other hand, the turbulent flow data could be reconciled (within uncertainties) with the Dittus-Boelter’s correlation, once the temperature dependence of viscosity was included in the prediction of the Reynolds number. While this finding is plausible, it could not be directly confirmed, because most papers do not report information about the temperature dependence of the viscosity for their nanofluids.

Thesis Supervisor: Jacopo Buongiorno
Title: Associate Professor of Nuclear Science and Engineering
ACKNOWLEDGEMENTS

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My sincere thanks also goes to my fellow research group mates for their valuable contributions and suggestions for my research work and also coordinating with me in various research related events. I would like to thank Eric Forrest, Hyungdae Kim, Bao Truong, Bren Phillips and Vivek Inder Sharma.

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Last but not the least, I would like to thank my family: my father Pramod Kumar Varshney, sister Priyanka Prabhat and good friend Manisha Nadir for supporting me morally and spiritually.
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<thead>
<tr>
<th>Symbol</th>
<th>Unit</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\mu$</td>
<td>kg/(s.m)</td>
<td>Viscosity of nanofluids</td>
</tr>
<tr>
<td>$k$</td>
<td>W/(m.K)</td>
<td>Thermal Conductivity of nanofluids</td>
</tr>
<tr>
<td>$\rho$</td>
<td>kg/m$^3$</td>
<td>Density of nanofluids</td>
</tr>
<tr>
<td>$C_p$</td>
<td>J/(kg.K)</td>
<td>Specific heat of nanofluid</td>
</tr>
<tr>
<td>$\mu_f$</td>
<td>kg/(s.m)</td>
<td>Viscosity of basefluid</td>
</tr>
<tr>
<td>$k_f$</td>
<td>W/(m.K)</td>
<td>Thermal conductivity of basefluid</td>
</tr>
<tr>
<td>$C_{pf}$</td>
<td>J/(kg.K)</td>
<td>Specific heat of basefluid</td>
</tr>
<tr>
<td>$k_p$</td>
<td>W/(m.K)</td>
<td>Thermal Conductivity of nanoparticles</td>
</tr>
<tr>
<td>$\rho_p$</td>
<td>kg/m$^3$</td>
<td>Density of nanoparticles</td>
</tr>
<tr>
<td>$C_{pp}$</td>
<td>J/(kg.K)</td>
<td>Specific heat of nanoparticles</td>
</tr>
<tr>
<td>$h$</td>
<td>W/m$^2$K</td>
<td>Heat Transfer Coefficient</td>
</tr>
<tr>
<td>$D$</td>
<td>m</td>
<td>Diameter of tube</td>
</tr>
<tr>
<td>$L$</td>
<td>m</td>
<td>Length of tube</td>
</tr>
<tr>
<td>$\phi$</td>
<td>dimensionless</td>
<td>Volume fraction of nanoparticle</td>
</tr>
<tr>
<td>$Nu$</td>
<td>dimensionless</td>
<td>Nusselt number</td>
</tr>
<tr>
<td>$Re$</td>
<td>dimensionless</td>
<td>Reynolds number</td>
</tr>
<tr>
<td>$Pe$</td>
<td>dimensionless</td>
<td>Peclet number</td>
</tr>
</tbody>
</table>
1. Introduction

Nanofluids are engineered colloidal dispersions of nanoparticles in base fluids such as water, oils or refrigerants. The nanoparticles can be metals such as copper, silver, gold or metal oxides such as alumina, zirconia, silica or various forms of carbon such as diamond, carbon nanotubes, graphite etc.

One of the advantages of using nanoparticles over micro-sized particles is that they do not clog the system and are expected to more closely mirror the behavior of molecules of the fluid. Erosion effects and settling will also decrease with the use of nanoparticles instead of micro-sized particles. Nanofluids research to date has examined fluids with particles of various types, including chemically stable metals, metal oxides, and carbon [1].

The most important factor which has called upon the attention of engineering community towards nanofluids is their improved heat transfer characteristics over base fluids. Nanofluids were first named and proposed as a new method of enhancing heat transfer in fluid-cooled systems by Choi in 1995 [2]. Even at very low concentration of nanoparticles, some believe that, nanofluids can show very high enhancement in the heat transfer capabilities with respect to the basefluid. This may make them useful to many practical applications involving fluid based heat transfer like cooling of automobiles, electronic circuits and transformers.

In spite of the attention received by this field, uncertainties concerning the fundamental effects of nanoparticles on thermo-physical properties of solvent media remain. Thermal conductivity is the property that has catalyzed the attention of the nanofluids research community the most. As dispersions of solid particles in a continuous liquid matrix, nanofluids are expected to have a thermal conductivity that obeys the effective medium theory developed by Maxwell over 100 years ago. Maxwell’s model for spherical and well-dispersed particles culminates in a simple equation giving the ratio of the nanofluid thermal conductivity ($k$) to the thermal conductivity of the basefluid ($k_b$):
where $k_p$ is the particle thermal conductivity and $\phi$ is the particle volumetric fraction. Note that the model predicts no explicit dependence of the nanofluid thermal conductivity on the particle size or temperature. Also, in the limit of $k_p \gg k_f$ and $\phi << 1$, the dependence on particle loading is expected to be linear, as given by $\frac{k}{k_f} \approx 1 + 3\phi$. However, several deviations from the predictions of Maxwell’s model have been reported, including:

- A strong thermal conductivity enhancement beyond that predicted by Eq. (1) with a non-linear dependence on particle loading \cite{4-10}
- A dependence of the thermal conductivity enhancement on particle size and shape \cite{9,11-19}
- A dependence of the thermal conductivity enhancement on fluid temperature \cite{14,20-22}

To explain these unexpected and intriguing findings, several hypotheses were recently formulated. For example, it was proposed that:

- Particle Brownian motion agitates the fluid, thus creating a micro-convection effect that increases energy transport \cite{23-27}
- Clusters or agglomerates of particles form within the nanofluid, and heat percolates preferentially along such clusters \cite{28-33}
- Basefluid molecules form a highly-ordered high-thermal-conductivity layer around the particles, thus augmenting the effective volumetric fraction of the particles \cite{28,32,34,35}

Experimental confirmation of these mechanisms has been weak; some mechanisms have been openly questioned. For example, the micro-convection hypothesis has been shown to yield predictions in conflict with the experimental evidence \cite{19,36}. In addition to theoretical inconsistencies, the nanofluid thermal conductivity data are sparse and inconsistent, possibly due to (i) the broad range of experimental approaches that have been implemented to measure nanofluid thermal conductivity (e.g., transient hot
wire, steady-state heated plates, oscillating temperature, thermal lensing), (ii) the often-incomplete characterization of the nanofluid samples used in those measurements, and (iii) the differences in the synthesis processes used to prepare those samples, even for nominally similar nanofluids. In summary, the possibility of very large thermal conductivity enhancement in nanofluids beyond Maxwell’s prediction and the associated physical mechanisms are still a hotly debated topic.

There have also been several experimental studies which have shown that adding nanoparticles to a fluid enhances the convective heat transfer. A literature search was conducted for publications on nanofluids convective heat transfer; the search yielded over 40 journal papers [54-103]. Most of the published studies report that addition of nanoparticles to base fluids enhances the convective heat transfer capabilities. Not all of these studies, however, take into account the change in the properties of the base fluids due to the addition of nanoparticles when predicting the behavior of nanofluids, which is then compared to the experimental values. Some studies [93-103] have reported abnormally high enhancements in the convective heat transfer of nanofluids which could not be predicted by the conventional heat transfer correlations. The research conducted at MIT [54-55], on the other hand, shows that if the properties of the nanofluids are properly accounted for in calculating the governing dimensionless numbers (Re, Pr and Nu), the existing correlations for convective heat transfer accurately predict the heat transfer coefficient enhancements seen with nanofluids. In summary, the nanofluid convective heat transfer database would seem to be inconsistent, and the question of whether truly abnormal convective heat transfer enhancement is achievable with nanofluids remains open.

Some experimental studies report enhancements in convective heat transfer which could not be explained by existing correlations, such as the Dittus-Boelter correlation for convective heat transfer in fully-developed turbulent flow:

\[
\begin{align*}

Nu &= 0.023 \text{Re}^{0.8} \text{Pr}^{0.4} \\

h &= 0.023 \frac{\text{Re}^{0.8} \cdot \text{Pr}^{0.4} \cdot \mu^{0.4}}{D} = 0.023 \frac{\text{Re}^{0.8} \cdot \text{k}^{0.6} \cdot \text{D}^{0.8} \cdot \text{v}^{0.8}}{\mu^{0.4} \cdot D^{0.2}}.
\end{align*}
\]
where, $h, \rho, \mu$ and $k$ are the convective heat transfer coefficient, density, viscosity and thermal conductivity of the nanofluid respectively. $D$ and $V$ are the diameter of the pipe and the velocity of the nanofluid respectively. Similar correlation for constant heat flux laminar flow which reproduces the complex analytical solution for local Nusselt number to within 1% [57]:

\[
\begin{align*}
\text{Nu} &= 1.302(x^+)^{-1/3} - 0.5 & x^+ &\leq 0.0015 \\
\text{Nu} &= 4.364 + 0.263(x^+)^{0.506} e^{-41(x^+/2)} & x^+ &> 0.0015
\end{align*}
\]

(3)

where, $\text{Nu} = \frac{hD}{k}$ and the dimensionless distance, $x^+ = \frac{(x / D)}{\text{Re Pr}}$

In light of the above considerations, the studies of thermal conductivity and convective heat transfer coefficient of nanofluids are of great interest to us. They have been discussed separately in Chapter-2 and Chapter-3. The final conclusions have been given in the conclusion chapter at the end of the thesis.
2. Analysis of Anomalous Thermal Conductivity Enhancement

At the first scientific conference centered on nanofluids (*Nanofluids: Fundamentals and Applications*, September 16-20, 2007, Copper Mountain, Colorado), it was decided to launch an international nanofluid property benchmark exercise (INPBE), to resolve the inconsistencies in the database and help advance the debate on nanofluid properties. As a part of the INPBE, 31 organizations around the world measured properties of standard nanofluid samples that were manufactured and shipped to them by a supplier. MIT led the effort and part of this thesis project was devoted to supporting INPBE. Therefore, this thesis dissertation reports on the INPBE effort on the thermal conductivity data.

2.1. Methodology

The exercise’s main objective was to compare thermal conductivity data obtained by different organizations for the same samples. Four sets of test nanofluids were procured (see Section 2.2). To minimize spurious effects due to nanofluid preparation and handling, all participating organizations were given identical samples from these sets, and were asked to adhere to the same sample handling protocol. The main points of the protocol were:

1. Complete all measurements within one month of the delivery date.
2. Store all samples in a cool, dry and dark environment until the measurements are completed.
3. Do not sonicate the samples or mix the samples by other means.
4. Do not add surfactants or other chemicals to the samples.

The exercise was ‘semi-blind’, as only minimal information about the samples was given to the participants at the time of sample shipment. The minimum requirement to participate in the exercise was to measure and report the thermal conductivity of at least one test nanofluid at room temperature. However, participants could also measure (at their discretion) thermal conductivity at higher temperature and/or various other nanofluid properties, including (but not necessarily limited to) viscosity, density,
specific heat, particle size and concentration. The data were then reported in a standardized form to the exercise coordinator at the Massachusetts Institute of Technology (MIT) and posted, unedited, at the INPBE website (http://mit.edu/nse/nanofluids/benchmark/index.html). The complete list of organizations that participated in INPBE, along with the data they contributed, is reported in Table I. INPBE climaxed in a workshop, held on January 29-30 in Beverly Hills, California, where the results were presented and discussed by the participants. The workshop presentations can also be found at the INPBE website.
<table>
<thead>
<tr>
<th>Organization / Contact person</th>
<th>Experimental method&lt;sup&gt;a&lt;/sup&gt; for thermal conductivity measurement (Ref.)</th>
<th>Generated data for</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Argonne National Laboratory / E. V. Timofeeva</td>
<td>KD2 Pro</td>
<td>TC&lt;sup&gt;b&lt;/sup&gt;</td>
<td>TC</td>
</tr>
<tr>
<td>CEA / C. Reynaud</td>
<td>Steady-state coaxial cylinders&lt;sup&gt;c&lt;/sup&gt;</td>
<td>TC</td>
<td></td>
</tr>
<tr>
<td>Chinese University of Hong Kong / S.-Q. Zhou</td>
<td>Steady state parallel plate&lt;sup&gt;d&lt;/sup&gt;</td>
<td>V&lt;sup&gt;e&lt;/sup&gt;</td>
<td>TC, V</td>
</tr>
<tr>
<td>DSO National Laboratories / L. G. Kieng</td>
<td>Supplied nanofluid samples</td>
<td></td>
<td></td>
</tr>
<tr>
<td>ETH Zurich and IBM Research / W. Escher</td>
<td>THW and parallel hot plates&lt;sup&gt;f&lt;/sup&gt;</td>
<td>TC</td>
<td>TC</td>
</tr>
<tr>
<td>Helmut-Schmidt University Armed Forces / S. Kabelac</td>
<td>Guarded hot plate&lt;sup&gt;d&lt;/sup&gt;</td>
<td>TC, V</td>
<td>TC, V</td>
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<td>Illinois Institute of Technology / D. Venerus</td>
<td>Forced Rayleigh scattering&lt;sup&gt;g&lt;/sup&gt;</td>
<td>TC, V</td>
<td>TC</td>
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<tr>
<td>Indian Institute of Technology, Kharagpur / I. Manna</td>
<td>KD2 Pro</td>
<td>TC</td>
<td>TC</td>
</tr>
<tr>
<td>Indian Institute of Technology, Madras / T. Sundararajan, S. K. Das</td>
<td>THW&lt;sup&gt;h&lt;/sup&gt;</td>
<td>TC</td>
<td>TC</td>
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<td>Institution</td>
<td>THW, KD2 Pro</td>
<td>TC</td>
<td>TC</td>
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<td>-------------------------------------------------</td>
<td>---------------</td>
<td>----</td>
<td>----</td>
</tr>
<tr>
<td>Indira Gandhi Centre for Atomic Research / J. Philip</td>
<td>THW^d, KD2</td>
<td>TC</td>
<td>TC</td>
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<td>Kent State University / Y. Tolmachev</td>
<td>KD2 Pro</td>
<td>TC</td>
<td>TC, V</td>
</tr>
<tr>
<td>Korea Aerospace University / S. P. Jang</td>
<td>THW^f</td>
<td>TC</td>
<td>TC</td>
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<tr>
<td>Korea Univ. / C. Kim</td>
<td>THW^d</td>
<td>TC, V</td>
<td>TC, V</td>
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<td>METSS Corp. / F. Botz</td>
<td>THW^d</td>
<td>TC</td>
<td>TC</td>
</tr>
<tr>
<td>MIT / J. Buongiorno, L. W. Hu, T. McKrell</td>
<td>THW^j</td>
<td>TC</td>
<td>TC</td>
</tr>
<tr>
<td>MIT / G. Chen</td>
<td>THW^k</td>
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<td>TC, V</td>
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<td>THW^l</td>
<td>TC</td>
<td>TC</td>
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<td>NIST / M. A Kedzierski</td>
<td>KD2 Pro</td>
<td>TC, V, D_m</td>
<td>V, D</td>
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<tr>
<td>North Carolina State University - Raleigh / J. Eapen</td>
<td>Contributed to data analysis</td>
<td></td>
<td></td>
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<td>Olin College of Engineering / R. Christianson, J. Townsend</td>
<td>THW^n</td>
<td>TC, V</td>
<td>TC</td>
</tr>
<tr>
<td>Queen Mary University of London / D. Wen</td>
<td>THW^d</td>
<td>TC, V</td>
<td>TC</td>
</tr>
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</table>
| Institution / Collaborators | Contribution | Equipment/Technique
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>RPI / P. Keblinski</td>
<td>Contributed to data analysis</td>
<td></td>
</tr>
<tr>
<td>SASOL of North America / Y. Chang</td>
<td>Supplied nanofluid samples</td>
<td></td>
</tr>
<tr>
<td>Silesian University of Technology / A. B. Jarzebski, G. Dzido</td>
<td>THW °</td>
<td>TC, V</td>
</tr>
<tr>
<td>South Dakota School of Mines and Technology / H. Hong</td>
<td>Hot Disk ‹</td>
<td>TC, TC, TC</td>
</tr>
<tr>
<td>Stanford University / P. Gharagozloo, K. Goodson</td>
<td>IR thermometry ‡</td>
<td>TC, TC</td>
</tr>
<tr>
<td>Texas A&amp;M University / J. L. Alvarado</td>
<td>KD2 Pro</td>
<td>TC, TC, TC</td>
</tr>
<tr>
<td>Ulsan National Institute of Science and Technology; Tokyo Institute of Technology / I. C. Bang, J. H. Kim</td>
<td>KD2 Pro</td>
<td>TC, V, TC, V, TC, V</td>
</tr>
<tr>
<td>Université Libre de Bruxelles, University of Naples / C. S. Iorio</td>
<td>Modified hot wall technique †, Parallel plates §</td>
<td>TC, V, D, TC, V, D</td>
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<td>University of Leeds / Y. Ding</td>
<td>KD2 and parallel hot plates †</td>
<td>TC, V, TC, V, TC, V</td>
</tr>
<tr>
<td>University of Pittsburgh / M. K. Chyu</td>
<td>Unitherm™ 2022 (Guarded heat flow meter)</td>
<td>TC, TC, TC, V</td>
</tr>
</tbody>
</table>
a THW = transient hot wire; KD2 and KD2 Pro (information about these devices at http://www.decagon.com/thermal/instrumentation/instruments.php); Unitherm™ 2022 (information about this device at http://anter.com/2022.htm)

b TC = thermal conductivity


d A publication with detailed information about this apparatus is not available

e V = viscosity


m D = density


Table II Characteristics of the Set 1 samples

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Loading</th>
<th>Particle size</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Sasol</td>
</tr>
<tr>
<td>1</td>
<td>1% vol.</td>
<td>1.2 to 1.3 % vol</td>
</tr>
<tr>
<td>2</td>
<td>n/a</td>
<td>n/a</td>
</tr>
<tr>
<td>3</td>
<td>1% vol.</td>
<td>0.7 to 0.8 % vol</td>
</tr>
<tr>
<td>4</td>
<td>3% vol.</td>
<td>1.9 to 2.2 % vol</td>
</tr>
<tr>
<td>5</td>
<td>1% vol.</td>
<td>0.7 to 0.8 % vol</td>
</tr>
<tr>
<td>6</td>
<td>3% vol.</td>
<td>2.0 to 2.3 % vol</td>
</tr>
<tr>
<td>7</td>
<td>n/a</td>
<td>n/a</td>
</tr>
</tbody>
</table>

Range of values is given to account for expected hydration range of alumina (boehmite). Boehmite’s chemical formula is Al₂O₃·nH₂O, where n = 1 to 2. The hydrate is bound and cannot be dissolved in water. In most boehmites there is 70 to 82 wt% Al₂O₃ per gram of powder. Boehmite density is 3.04 g/cm³.
Average size of dispersed phase, measured by dynamic light scattering (DLS). The range indicates the spread of six nominally-identical measurements. DLS systemic uncertainty is of the order of ±10 nm. Malvern NanoS used to collect data.

Measurements by inductive coupled plasma (ICP)

Average size of dispersed phase, measured by DLS. The range indicates the spread of multiple nominally-identical measurements. DLS systemic uncertainty is of the order of ±10 nm.

Not applicable

Measurements by neutron activation analysis (NAA)

Not available due to unreliability of DLS analyzer with PAO-based samples


<table>
<thead>
<tr>
<th>Sample #</th>
<th>Au loading</th>
<th>Particle size</th>
<th>Stabilizer concentration (trisodium citrate)</th>
<th>pH</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>DSO</td>
<td>MIT</td>
</tr>
<tr>
<td>1</td>
<td>0.0010 vol%</td>
<td>0.0009 vol%</td>
<td>20-30 nm</td>
<td>4-11 nm</td>
</tr>
<tr>
<td>2</td>
<td>Zero</td>
<td>Zero</td>
<td>n/a</td>
<td>n/a</td>
</tr>
</tbody>
</table>

Measurements by inductive coupled plasma (ICP). ICP has an accuracy of 0.6% of the reported value for gold in the concentration range of interest.

Number-weighted average size of particles, measured by DLS. The range indicates the spread of two nominally-identical measurements.

DLS systemic uncertainty is of the order of ±10 nm.

Measurements by DLS. The values reported are the number-weighted average and the range at the full-width half maximum for six measurements.
Assumed density of gold is $19.32 \text{ g/cm}^3$

Within the detection limit of ICP.

Not applicable

**Table IV Characteristics of the Set 3 samples**

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Silica (SiO$_2$) loading</th>
<th>Na$_2$SO$_4$ concentration</th>
<th>Particle size</th>
<th>pH</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Grace</td>
<td>MIT</td>
<td>Grace</td>
<td>MIT</td>
</tr>
<tr>
<td>1</td>
<td>49.8 wt%</td>
<td>43.6 wt%</td>
<td>0.1-0.2 wt% of Na</td>
<td>0.27 wt% of Na</td>
</tr>
<tr>
<td></td>
<td>31.1 vol%</td>
<td>26.0 vol%</td>
<td>Na</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>n/a</td>
<td>n/a</td>
<td>n/a</td>
<td>n/a</td>
</tr>
</tbody>
</table>

*a* Measured by inductive coupled plasma (ICP). ICP has an accuracy of 0.6% of the reported value.

*b* Number-weighted average size of particles, measured by dynamic light scattering (DLS). The range indicates the spread of three nominally-identical measurements. DLS systemic uncertainty is of the order of ±10 nm.

*c* Assumed density of silica (SiO$_2$) is 2.2 g/cm$^3$

*d* Sample 2 is simply de-ionized water, which was assumed to be the basefluid sample, but was not actually sent to the participants.
### Table V Characteristics of the Set 4 samples.

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Particle loading</th>
<th>Particle composition</th>
<th>Particle size</th>
<th>pH</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>UPRM</td>
<td>MIT</td>
<td>UPRM</td>
<td>MIT</td>
</tr>
<tr>
<td>1</td>
<td>0.17 vol%(^b)</td>
<td>0.16 vol%(^c)</td>
<td>Mn(<em>{32})-Zn(</em>{32})-Fe(_2)(^d)</td>
<td>Mn ~ 15 at%, Zn ~ 14 at%, Fe ~ 71 at%</td>
</tr>
<tr>
<td>2</td>
<td>n/a(^g)</td>
<td>n/a</td>
<td>n/a</td>
<td>n/a</td>
</tr>
</tbody>
</table>

---

\(^a\) Atomic fraction of metals measured by Energy Dispersive X-ray Spectroscopy (EDS).

\(^b\) Determined from magnetic measurements

\(^c\) Measurements by inductive coupled plasma (ICP). Assumed density of 4.8 g/cm\(^3\) for Mn-Zn ferrite. ICP has an accuracy of 0.6% of the reported value.

\(^d\) The molar fraction of Mn and Zn was determined from stoichiometric balance.

\(^e\) Average magnetic particle diameter
Number-weighted average size of particles, measured by dynamic light scattering (DLS). The range indicates the spread of four nominally-identical measurements. DLS systemic uncertainty is of the order of ±10 nm.

**Not applicable**

---

**Table VI Summary of INPBE results**

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Sample description</th>
<th>Measured thermal conductivity (W/m-K)</th>
<th>Measured thermal conductivity ratio</th>
<th>Predicted thermal conductivity ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>[Formula]</td>
<td>[Formula]</td>
<td>[Formula]</td>
</tr>
<tr>
<td>Sample 1</td>
<td>Alumina nanorods (80x10 nm), 1% vol. in water</td>
<td>0.627 ± 0.013</td>
<td>1.036 ± 0.004</td>
<td>1.024</td>
</tr>
<tr>
<td>Sample 2</td>
<td>De-ionized water</td>
<td>0.609 ± 0.003</td>
<td>n/a</td>
<td>n/a</td>
</tr>
<tr>
<td>Sample 3</td>
<td>Alumina nanoparticles (10 nm), 1% vol. in PAO + surfactant</td>
<td>0.162 ± 0.004</td>
<td>1.039 ± 0.003</td>
<td>1.027</td>
</tr>
<tr>
<td>Sample 4</td>
<td>Alumina nanoparticles (10 nm), 3% vol. in PAO + surfactant</td>
<td>0.174 ± 0.005</td>
<td>1.121 ± 0.004</td>
<td>1.083</td>
</tr>
<tr>
<td>Sample</td>
<td>Description</td>
<td>Value 1</td>
<td>Value 2</td>
<td>Value 3</td>
</tr>
<tr>
<td>--------</td>
<td>------------------------------------------------------------------------------</td>
<td>---------</td>
<td>---------</td>
<td>---------</td>
</tr>
<tr>
<td>Sample 5</td>
<td>Alumina nanorods (80x10 nm), 1% vol. in PAO + surfactant</td>
<td>0.164 ± 0.005</td>
<td>1.051 ± 0.003</td>
<td>1.070</td>
</tr>
<tr>
<td>Sample 6</td>
<td>Alumina nanorods (80x10 nm), 3% vol. in PAO + surfactant</td>
<td>0.182 ± 0.006</td>
<td>1.176 ± 0.005</td>
<td>1.211</td>
</tr>
<tr>
<td>Sample 7</td>
<td>PAO + surfactant</td>
<td>0.156 ± 0.005</td>
<td>n/a</td>
<td>n/a</td>
</tr>
<tr>
<td>Set 2</td>
<td>Sample 1 Gold nanoparticles (10 nm), 0.001% vol. in water + stabilizer</td>
<td>0.613 ± 0.005</td>
<td>1.007 ± 0.003</td>
<td>1.000</td>
</tr>
<tr>
<td></td>
<td>Sample 2 Water + stabilizer</td>
<td>0.604 ± 0.003</td>
<td>n/a</td>
<td>n/a</td>
</tr>
<tr>
<td>Set 3</td>
<td>Sample 1 Silica nanoparticles (22 nm), 31% vol. in water + stabilizer</td>
<td>0.729 ± 0.007</td>
<td>1.204 ± 0.010</td>
<td>1.008</td>
</tr>
<tr>
<td></td>
<td>Sample 2 De-ionized water</td>
<td>0.604 ± 0.002</td>
<td>n/a</td>
<td>n/a</td>
</tr>
<tr>
<td>Set 4</td>
<td>Sample 1 Mn-Zn ferrite nanoparticles (7 nm), 0.17% vol. in water + stabilizer</td>
<td>0.459 ± 0.005</td>
<td>1.003 ± 0.008</td>
<td>1.000</td>
</tr>
<tr>
<td></td>
<td>Sample 2 Water + stabilizer</td>
<td>0.455 ± 0.005</td>
<td>n/a</td>
<td>n/a</td>
</tr>
</tbody>
</table>

- **Nominal values for nanoparticle concentration and size**
- **Sample average and standard error of the mean**
- **Calculated with the assumptions in Appendix B**
- **Not applicable**
2.2. **Test nanofluids**

To strengthen the generality of the INPBE results, it was desirable to select test nanofluids with a broad diversity of parameters; for example, we wanted to explore aqueous and non-aqueous basefluids, metallic and oxidic particles, near-spherical and elongated particles, and high and low particle loadings. Also, given the large number of participating organizations, the test nanofluids had to be available in large quantities (> 2 L) and at reasonably low cost.

Accordingly, four sets of test samples were procured. The providers were Sasol (Set 1), DSO National Labs (Set 2), W. R. Grace & Co. (Set 3) and the University of Puerto Rico at Mayaguez (Set 4). The providers reported information regarding the particle materials, particle size and concentration, basefluid material, the additives/stabilizers used in the synthesis of the nanofluid, and the material safety data sheets. Said information was independently verified, to the extent possible, by the INPBE coordinators (MIT and Illinois Institute of Technology, IIT), as reported in the next sections. Identical samples were shipped to all participating organizations.

2.2.1. **Set 1**

The samples in Set 1 were supplied by Sasol. The numbering for these samples is as follows:

1. Alumina nanorods in de-ionized water
2. De-ionized water. (basefluid sample)
3. Alumina nanoparticles (first concentration) in Polyalphaolefins lubricant (PAO) + surfactant
4. Alumina nanoparticles (second concentration) in PAO + surfactant
5. Alumina nanorods (first concentration) in PAO + surfactant
6. Alumina nanorods (second concentration) in PAO + surfactant
7. PAO + surfactant. (basefluid sample)
The synthesis methods have not been published, so a brief summary is given here. For sample 1, alumina nanorods were simply added to water and dispersed by sonication. Sample 2, de-ionized water, was not actually sent to the participants. The synthesis of samples 3-7 involved three steps. First, the basefluid was created by mixing PAO (SpectraSyn-10 by Exxon Oil) and 5% wt. dispersant (Solsperse 21000 by Lubrizol Chemical), and heating and stirring the mixture at 70°C for two hours, to ensure complete dissolution of the dispersant. Second, hydrophilic alumina nanoparticles or nanorods (in aqueous dispersion) were coated with a mono-layer of hydrophobic linear alkyl benzene sulfonic acid and then spray dried. Third, the dry nanoparticles or nanorods were dispersed into the basefluid.

![TEM pictures of samples 1 (a) and 3 (b), respectively.](image)

**Figure 1.** Set 1 - TEM pictures of samples 1 (a) and 3 (b), respectively. The nanorod dimensions in sample 1 are in reasonable agreement with the nominal size (80x10nm) stated by Sasol. However, smaller particles of lower aspect ratio are clearly present along with the nanorods. TEM pictures of PAO-based samples have generally been of lower quality. However, the nanoparticles in sample 3 appear to be roughly spherical and of approximate diameter 10-20 nm, thus consistent with the nominal size of 10 nm stated by Sasol.

Table II reports the information received by Sasol along with the results of some measurements done at MIT and IIT. Figure 1 shows transmission electron microscopy (TEM) images for samples 1 and 3. TEM images for samples 4-6 are not available.
2.2.2. Set 2

The samples in Set 2 were supplied by Dr. Lim Geok Kieng of DSO National Laboratories in Singapore. The numbering for these samples is as follows:

1. Gold nanoparticles in de-ionized water and trisodium citrate stabilizer.
2. De-ionized water + sodium citrate stabilizer. (basefluid sample)

![TEM pictures of sample 1](image_url)

**Figure 2.** Set 2 - TEM pictures of sample 1. The nanoparticles are roughly spherical and of diameter <20 nm, thus somewhat smaller than the nominal size of 20-30 nm stated by DSO National Labs.

The nanofluid sample was produced according to a one-step ‘citrate method’, in which 100 mL of 1.18 mM gold(III) chloride trihydrate solution and 10 mL of 3.9 M trisodium citrate dehydrate solution were mixed, brought to boil and stirred for 15 minutes. Gold nanoparticles formed as the solution was let cool to room temperature. Table III reports the information received by DSO National Labs along with the results of some measurements done at MIT. Figure 2 shows the TEM images for sample 1.
2.2.3. Set 3

Set 3 consisted of a single sample, supplied by W. R. Grace & Co.:

(1) Silica mono-dispersed spherical nanoparticles and stabilizer in de-ionized water

Figure 3. Set 3 - TEM pictures of sample 1. The nanoparticles are roughly spherical and of diameter 20-30 nm, thus consistent with the nominal size of 22 nm stated by Grace.

The silica particles were synthesized by ion exchange of sodium silicate solution in a proprietary process. A general description of this process can be found in the literature. Grace commercializes this nanofluid as Ludox TM-50, and indicated that the nanoparticles are stabilized by making the system alkaline, the base being deprotonated silanol (SiO⁻) groups on the surface with Na⁺ as the counterion (0.1-0.2 wt% of Na ions). The dispersion contains also 500 ppm of a proprietary biocide. Grace stated that it was not possible to supply a basefluid sample with only water and stabilizer “because of the way the particles are made”. Given the low concentration of the stabilizer and biocide, de-ionized water was assumed to be the basefluid sample, and designated ‘sample 2’, though it was not actually sent to the participants. Table IV
reports the information received by Grace along with the results of some measurements done at MIT. Figure 3 shows the TEM images for the Set 3 sample.

2.2.4. Set 4

The samples in Set 4 were supplied by Dr. Jorge Gustavo Gutierrez of the University of Puerto Rico – Mayaguez (UPRM). A chemical co-precipitation method was used to synthesize the particles\textsuperscript{41}. The Set 4 sample numbering is as follows:

(1) Mn-Zn ferrite (\(\text{Mn}_{x}\text{Zn}_{1-x}\text{Fe}_2\text{O}_4\)) particles in solution of stabilizer and water.

(2) Solution of stabilizer (25 wt %) and water (75 wt %). (basefluid sample)

\[\text{Figure 4. Set 4 - TEM pictures of sample 1. The nanoparticles have irregular shape and approximate size <20 nm, thus consistent with the nominal size of \(\sim 7\) nm stated by UPRM.}\]

The stabilizer is Tetramethylammonium hydroxide, or (CH\(_3\))\(_4\)NOH. Table V reports the information received by UPRM along with the results of some measurements done at MIT. Figure 4 shows the TEM images for sample 1.
2.3. Thermal conductivity data

The thermal conductivity data generated by the participating organizations are shown in Figures 5 through 17, one for each sample in each set. In these figures the data are anonymous, i.e., there is no correspondence between the organization number in the figures and the organization list in Table I. The data points indicate the mean value for each organization, while the error bars indicate the standard deviation calculated using the procedure described in Appendix A. The sample average, i.e., the average of all data points, is shown as a solid line, and the standard error of the mean is denoted by the dotted lines to facilitate visualization of the data spread. The standard error of the mean is typically much lower than the standard deviation because it takes into account the total number of measurements made to arrive at the sample average. Each measurement technique is denoted by a different symbol, and averages for each of the measurement techniques are shown. The measurement techniques were grouped into four categories: the KD2 Thermal Properties Analyzer (Decagon), custom transient hot wire (THW), steady state parallel plate, and other techniques. Outliers (determined using Peirce's criterion) are shown as filled data points and were not included in either the technique or ensemble averages.

It can be seen that for all water-based samples in all four sets most organizations report values of the thermal conductivity that are within ±5% of the sample average. For the PAO-based samples the spread is a little wider, with most organizations reporting values that are within ±10% of the sample average. A note of caution is in order: while all data reported here are nominally for room temperature, what constitutes 'room temperature' varies from organization to organization. The data shown in Figures 5 through 17 include only measurements conducted in the range 20 – 30 °C. Over this range of temperatures, the thermal conductivity of the test fluids is expected to vary minimally; for example, the water thermal conductivity varies by less than 2.5%. Where deionized water was the basefluid (Figures 6 and 15), the range of nominal thermal conductivity of water for 20 – 30 °C is shown as a red band plotted on top of the measured data.
Figures 18 through 25 show the thermal conductivity ‘enhancement’ for all nanofluid samples, i.e., the ratio of the nanofluid thermal conductivity to the basefluid thermal conductivity. For each organization, the data point represents the ratio of the mean thermal conductivities of the nanofluid and basefluid, while the error bars represent the standard deviation calculated according to the procedure described in Appendix A. If a participating organization did not measure the basefluid thermal conductivity in their laboratory, a calculation of enhancement was not made. Again, the sample average is shown as a solid line along with the standard error of the mean, and outliers are indicated by filled data points. Note that there is reasonable consistency (within ±5%) in the thermal conductivity ratio data among most organizations and for all four sets, including water-based and PAO-based samples.

The INPBE database is summarized in Table VI. Comparing the data for samples 3, 4, 5 and 6 in Set 1, it is noted that, everything else being the same, the thermal conductivity enhancement is higher at higher particle concentration, and higher for elongated particles than for near-spherical particles. Comparing the data for samples 1 and 5 in Set 1, it is noted that the thermal conductivity enhancement is somewhat higher for the PAO basefluid than for water. The Set 2 data suggest that the thermal conductivity enhancement is negligible, if the particle concentration is very low, even if metal particles of high thermal conductivity are used. On the other hand, the Set 3 data suggest that a robust enhancement can be achieved, if the particle concentration is high, even if the particle material has a modest thermal conductivity. All these trends are expected, based on the effective medium theory, as will be discussed in section 2.4.

2.3.1. Effects of the Experimental Approach on the Thermal Conductivity Measurements

Table I reports the experimental techniques used by the various organizations to measure thermal conductivity, and provides, when available, a reference where more information about those techniques can be found. Transient, steady-state, and optical techniques were used to measure thermal conductivity. There are transient measurement techniques that require the immersion of a dual heating and sensing element in the sample, such as the transient hot wire (THW) and transient hot disk techniques. The THW
The THW technique was used by over half of the participating organizations, many of which used a custom built apparatus. The KD2 Thermal Properties Analyzer made by Decagon, an off-the-shelf device that is based on the THW approach, was also used. The transient hot disk technique is similar to the THW technique, except that the heater/sensor is a planar disk coated in Kapton [40]. In steady-state techniques such as the parallel plate [41] and co-axial cylinder [42] methods, heat is transferred between two plates (or co-axial cylinders) sandwiching the test fluid. Measurement of the temperature difference and heat transfer rate across the fluid can be used to determine the thermal conductivity via Fourier's law. The thermal comparator method, also a steady-state method, measures the voltage difference between a heated probe in point contact with the surface of the fluid sample and a reference, which can be converted to thermal conductivity using a calibration curve of samples of known conductivity [43]. In the forced Rayleigh scattering method, an optical grating is created in a sample of the fluid using the intersection of two beams from a high-powered laser. The resulting temperature change causes small-scale density changes that create a refraction index grating that can be detected using another laser. The relaxation time of the refraction index grating is related to the thermal diffusivity of the fluid, from which the thermal conductivity can be evaluated [44].

The measurement techniques were grouped into KD2, Custom THW, Parallel Plate, and Other (which include thermal comparator, hot disk, Forced Rayleigh Scattering, and co-axial cylinders). Thermal conductivity and enhancement data for each group of measurement techniques is shown in Figures 5 – 25.

For each of the four measurement technique groupings, the average thermal conductivity is shown on the plot and is indicated by the solid line. In the custom THW data on Figure 5 and 6, there is one measurement that is well above the average in both figures. This was the only THW apparatus with an uninsulated wire. Typically an insulated wire is used in this method to reduce the current leakage into the fluid. The higher thermal conductivity measured here may be a result of that effect. Excepting the
outliers, all the measurement techniques show good agreement for deionized water (Figures 6 and 15). For the PAO basefluid (Figure 11) the uninsulated hot wire measurement (Organization 14) is no longer an outlier. PAO is not as electrically conductive as water, and the current leakage effect should be less of an issue for this fluid.

As described in Appendix A, a Fixed Effects Model was used to determine whether differences in the data from different measurement techniques are statistically significant. Because of the low number of data points in the Parallel Plate and Other categories, only the KD2 and Custom Hot Wire techniques were compared. For all the samples in Sets 1, 2 and 3, the KD2 thermal conductivity average is lower than the Custom THW average. The Fixed Effects Model shows that this is a statistically significant difference for samples 1, 3, 4, 6, 7, in Set 1, and sample 2 in Set 3. In Set 4, the KD2 average is higher than the Custom THW, but this difference is statistically significant only for Sample 2 (the water+stabilizer basefluid for the ferrofluid). It is not clear why the KD2 measurements are lower than the THW measurements for all fluids except those in Set 4. Finally, in most cases, there is less scatter in the KD2 data for the PAO-based nanofluids than the water-based nanofluids. This may be due to the higher viscosity of the PAO which counteracts thermal convection during the 30 second KD2 heating cycle.

It is difficult to make specific conclusions about thermal conductivity measurements using the parallel plate technique due to the low number of data points and the amount of scatter for some samples (see Figures 12 – 14). Additional measurements would be needed to determine if there is a systematic difference between the parallel plate technique and other techniques.

Although the thermal conductivity data show some clear differences in measurement technique, these differences become less apparent once the data are normalized with the basefluid thermal conductivities (Figures 18 – 25). A comparison of the KD2 and THW techniques was again performed using the Fixed Effects Model. The only statistically significant difference between the two techniques was for Set 1, Sample 4 (Figure 20), and the 3% volume fraction alumina-PAO nanofluid.
This study shows that the choice of measurement technique can affect the measured value of thermal conductivity, but if the enhancement is the parameter of interest, the measurement technique is less important, at least for the KD2 and THW techniques. Therefore, to ensure accurate determinations of nanofluid thermal conductivity enhancement using these techniques, it is important to measure both the basefluid and nanofluid thermal conductivity using the same technique and at the same temperature.

**Figure 5.** Thermal conductivity data for sample 1, Set 1.

**Figure 6.** Thermal conductivity data for sample 2 (basefluid), Set 1.
Figure 7. Thermal conductivity data for sample 3, Set 1.

Figure 8. Thermal conductivity data for sample 4, Set 1.
**Set 1: Sample 5**

![Graph of Thermal Conductivity for Sample 5, Set 1](image)

*Figure 9. Thermal conductivity data for sample 5, Set 1.*

**Set 1: Sample 6**

![Graph of Thermal Conductivity for Sample 6, Set 1](image)

*Figure 10. Thermal conductivity data for sample 6, Set 1.*
Figure 11. Thermal conductivity data for sample 7, Set 1.

Figure 12. Thermal conductivity data for sample 1, Set 2.
Figure 13. Thermal conductivity data for sample 2, Set 2.

Figure 14. Thermal conductivity data for sample 1, Set 3.
Figure 15. Thermal conductivity data for sample 2 (basefluid), Set 3.

Figure 16. Thermal conductivity data for sample 1, Set 4.
Figure 17. Thermal conductivity data for sample 2 (basefluid), Set 4.

Figure 18. Thermal conductivity enhancement data for sample 1, Set 1.
Figure 19. Thermal conductivity enhancement data for sample 3, Set 1.

Figure 20. Thermal conductivity enhancement data for sample 4, Set 1.
Figure 21. Thermal conductivity enhancement data for sample 5, Set 1.

Figure 22. Thermal conductivity enhancement data for sample 6, Set 1.
Figure 23. Thermal conductivity enhancement data for sample 1, Set 2.

Figure 24. Thermal conductivity enhancement data for sample 1, Set 3.
**Figure 25.** Thermal conductivity enhancement data for sample 1, Set 4.

**Figure 26.** Percentage of all INPBE experimental data that are predicted by Nan et al.'s theory within the error indicated on the x-axis.
2.4. Comparison of data to effective medium theory

Equation (1) is valid for well-dispersed non-interacting spherical particles with negligible thermal resistance at the particle/fluid interface. To include the effects of particle geometry and finite interfacial resistance, Nan et al. [45] generalized Maxwell's model to yield the following expression for the thermal conductivity ratio:

$$\frac{k}{k_f} = \frac{3 + \phi[2\beta_{11}(1-L_{11}) + \beta_{33}(1-L_{33})]}{3 - \phi(2\beta_{11}L_{11} + \beta_{33}L_{33})}$$

where, for particles shaped as prolate ellipsoids with principal axes $a_{11} = a_{22} < a_{33}$:

$$L_{11} = \frac{p^2}{2(p^2-1)} - \frac{p}{2(p^2-1)^{3/2}} \cosh^{-1} p$$

$$L_{33} = 1 - 2L_{11}$$

$$p = \frac{a_{33}}{a_{11}}$$

and $R_{bd}$ is the (Kapitza) interfacial thermal resistance. The limiting case of very long aspect ratio in Nan et al.‘s theory is bounded by the nanoparticle linear aggregation models proposed by Prasher et al. [31] and Keblinski et al. [46]. Obviously, Eq. (4) reduces to Eq. (1) for spherical particles ($p=1$) and negligible interfacial thermal resistance ($R_{bd}=0$), as it can be easily verified. Equation (4) predicts that, if $k_p > k_f$, the thermal conductivity enhancement increases with increasing particle loading, increasing particle aspect ratio and decreasing basefluid thermal conductivity, as observed for the data in INPBE Set 1. More quantitatively, the theory was applied to the INPBE test nanofluids with the assumptions reported in Appendix B. Figure 26 shows the cumulative accuracy information of the effective medium theory for all the INPBE data. Two curves are shown: one for zero interfacial thermal resistance (upper bound), and one for a typical value of the interfacial resistance, $10^{-8}$ m$^2$K/W [47-49]. It can be seen that all INPBE data can be predicted by the lower bound theory with <17% error, while the upper bound estimate predicts 90% of the data with <18% error.
The above data analysis demonstrates that our colloidally stable nanofluids exhibit thermal conductivity in good agreement with the predictions of the effective medium theory for well-dispersed nanoparticles. That is, no anomalous thermal conductivity enhancement was observed for the nanofluids tested in this study. As such, resorting to the other theories proposed in the literature (e.g., Brownian motion, liquid layering, aggregation) is not necessary for the interpretation of the INPBE database. It should be noted, however, that the ranges of parameters explored in INPBE, while broad, are not exhaustive. For example, only one nanofluid with metallic nanoparticles was tested, and only at very low concentration.

2.5. Comparison of data with other theoretical models

The data obtained during INPBE was also compared with other theoretical models (except the effective medium theory) which have been proposed based on different possible mechanisms of heat transfer. Since the main aim of the study was to create a benchmark of properties of certain nanofluids and see if a particular theoretical model is able to capture the effect of all the parameters which were varied across different nanofluids used in this study, a comparison to various different theoretical models was necessary. The theoretical models that were implemented can be broadly classified into three different categories:

1. Static Models (Effective Medium Theories for spherical and non-spherical particles)

   a. Maxwell’s model for spherical nanoparticles [3]

   \[
   \frac{k}{k_f} = \frac{1 + 2\beta \phi}{1 - \beta \phi}
   \]  \hspace{1cm} (5)

   where, \( \beta = \frac{[k]}{(k_p + 2k_f)} \) and \([k] = k_p - k_f\)

   b. Maxwell’s model for non-spherical nanoparticles (Nan et al [45])

   \[
   \frac{k}{k_f} = \frac{3 + \phi[2\beta_{11}(1-L_{11}) + \beta_{33}(1-L_{33})]}{3 - \phi(2\beta_{11}L_{11} + \beta_{33}L_{33})}
   \]  \hspace{1cm} (6)
where, for particles shaped as prolate ellipsoids with principal axes $a_{11} = a_{22} < a_{33}$:

\[
L_{11} = \frac{p^2}{2(p^2 - 1)} - \frac{p}{2(p^2 - 1)^{3/2}} \cosh^{-1} p, \quad L_{33} = 1 - 2L_{11}, \quad p = a_{33} / a_{11}
\]

\[
\beta_{ii} = \frac{k_{ii}^c - k_f}{k_f + L_{ii}(k_{ii}^c - k_f)} , \quad k_{ii}^c = \frac{k_p}{1 + \gamma L_{ii} k_p / k_f} , \quad \gamma = (2 + 1/p) R_b k_f / (a_{11}/2)
\]

2. **Dynamic Models:** These models assume that there is a significant contribution of motion of nanoparticles which produces convection effect enhancing the thermal conduction in nanofluids.

a. **Prasher’s Dynamic Model** [25]

\[
\frac{k}{k_f} = (1 + A Re^{m Pr^{0.333}}) \left( \frac{[k_p (1 + 2\alpha) + 2k_f] + 2\phi [k_p (1 - \alpha) - k_f]}{[k_p (1 + 2\alpha) + 2k_f] - \phi [k_p (1 - \alpha) - k_f]} \right)
\]  

(7)

where, $A$ is a constant with value $4 \times 10^4$, $m$ is 2.0 for water based nanofluids

\[
\alpha = \frac{2 R_b k_f}{d} \quad \text{where} \quad R_b \text{ is the interfacial surface resistance and} \quad d \text{ is the diameter of nanoparticle. The value of} \quad R_b \text{ was chosen to be} \quad 1 \times 10^{-8} \text{ and} \quad 0 \text{ for predicting the lower and upper limit of thermal conductivities respectively.}
\]

b. **Jang and Choi Model** [7]

\[
k = k_f (1 - \phi) + k_{nano} \phi + 3 C_1 \frac{d_f}{d_p} k_f Re_d^2 Pr \phi
\]

(8)

\[
k_{nano} = \beta k_p
\]

where, $C_1$ is proportionality constant, $d_f$ and $d_p$ are diameter of basefluid molecule and nanoparticles respectively and $Re_d = \frac{C_{RM} d_p}{\nu_f}$ where $C_{RM}$ and $\nu_f$ are the random motion velocity of nanoparticles and dynamic viscosity of basefluid respectively. $C_{RM} = D_o / l_f$ where $D_o$ and $l_f$ are the
nanoparticle diffusion coefficient and mean free path in basefluid. \( D_0 = \frac{k_b T}{3\pi \mu d_p} \) where \( k_b \) is the Boltzmann constant. Mean free path was calculated using kinetic theory of gases. The value of \( C_f \) was assumed to be 510000 and \( \beta = 0.01 \)

3. **Aggregation Models**: These models assume an increase in the conduction due to aggregation of nanoparticles in the fluid forming larger clusters which lead to high conduction

a. Prasher's Aggregate Model [31]

\[
\frac{k}{k_f} = \frac{(k_a + 2k_f) + 2\varnothing_a(k_a - k_f)}{(k_a + 2k_f) - \varnothing_a(k_a - k_f)}
\]

where, \( k_a \) and \( \varnothing_a \) are the effective thermal conductivity and volume fraction of aggregates respectively.

\[
k_a = k_nc \left( \frac{3 + \varnothing_c [2\varnothing_c (1 - L_{11}) - \beta_{33} (1 - L_{33})]}{3 - \varnothing_c [2\varnothing_c L_{11} + \beta_{33} L_{33}]} \right)
\]

where, \( k_nc \) is the effective thermal conductivity of the aggregate sphere in the presence of the dead-end particles only. \( k_nc \) is given by the Bruggeman model as given below:

\[
\frac{(1 - \varnothing_{nc})(k_f - k_{nc})}{k_f + 2k_{nc}} + \frac{\varnothing_{nc}(k_p - k_{nc})}{k_f + 2k_{nc}} = 0
\]

where, \( \varnothing_{nc} \) is the volume fraction of particles with dead ends.

\[
L_{11} = \frac{p^2}{2(p^2 - 1)} - \frac{p}{2(p^2 - 1)^{3/2}} \cosh^{-1} p, \quad L_{33} = 1 - 2L_{11}, \quad p = R_g / \alpha
\]

\[
\beta_{11} = \frac{k_{nc} - k_f}{k_f + L_{11}(k_{nc} - k_f)} \quad \beta_{33} = \frac{k_{nc} - k_f}{k_f + L_{33}(k_{nc} - k_f)}
\]

\[
\varnothing_a = \varnothing_p / \varnothing_{int} \quad \varnothing_{nc} = \varnothing_{int} - \varnothing_c \quad \left( \frac{R_g}{\alpha} \right)_{max} = (\varnothing_p)^{1/(d_f - 3)}
\]
where, $\phi_{int}$ and $\phi_p$ are the volume fraction of nanoparticles in the aggregate/cluster and nanofluid respectively. $R_g$ and $a$ are the radii of the cluster and nanoparticles respectively. The aggregation parameter $p = (R_g/a)$ was varied from a minimum value of 2 to a maximum value of $(\phi_p)^{1/(d_f-3)}$ to obtain prediction of minimum and maximum thermal conductivity.

Section 2.4 discusses the comparison of INPBE data with the static models only. Plots shown in Figure 27 and 28 represent the comparison of the all the experimental data obtained during INPBE with the Maxwell Model for spherical particles. Figure 2 shows that 90% of the data measured from experiments was predicted by the Maxwell’s model for spherical particles within 10% accuracy Figure 28 confirms the same fact in a different way, it shows the comparison of theoretical estimation and experimental measurements with 10% error bars and it is clear from the plot that most of the data is predicted within 10% of the measured values using the traditional Maxwell’s model for predicting thermal conductivity.

A similar comparison was done with the modified Maxwell’s model given by Nan et al [45] which takes into account the non-spherical particles like nanorods. The comparison of the experimental measurements with the estimation from the Nan et al have been discussed in Section 2.4. Hence in general the traditional static thermal conductivity models seem to predict the thermal conductivity data was also compared with dynamic and aggregation models mentioned above. Dynamic models analyzed for the INPBE samples included Prasher’s Dynamic Model and Jang and Choi’s dynamic Model. These models assume that the motion of nanoparticles in a nanofluid is responsible for the enhancement of thermal conductivity. A comparison of the experimental results obtained during INPBE with the Prasher’s Dynamic model has been shown in Figure 29 and Figure 30. It is clear from the plots that only 30% of the experimental results were predicted within 10% accuracy by Prasher’s Dynamic model.

The comparison with Jang and Choi’s Dynamic model is shown in Figure 31 and Figure 32. Figure 31 suggests that the model highly over predicts the results for certain experiments. These were found to be
samples with high concentration of nanoparticles. Figure 32 shows that around 70% of the experimental values are predicted by the Jang and Choi's model within 10% experimental error. Hence it has been observed that dynamic models in general seem to over predict the thermal conductivity of nanofluids especially at higher concentration of nanoparticles.
Figure 27: Percentage of result plotted as a function of Fractional Error for Maxwell's Static model for spherical particles.

Figure 28: Calculated Value from the Maxwell Static model is plotted against the measured values with 10% error bars.
Figure 29 Percentage of results plotted as a function of fractional error for Prasher's Dynamic Model

Figure 30 Calculated value from Prasher's Dynamic model vs Measured Values with 10% error bars
Figure 31 Calculated value from Jang and Choi's Dynamic model vs Measured Values with 10% error bars

Figure 32 Percentage of results plotted as a function of fractional error for Jang and Choi's Dynamic Model
Figure 33 Calculated value from Prasher's Aggregate model vs Measured Values with 10% error bars, blue dots represent the upper limit and red dots represent the lower limit.

Figure 34 Percentage of results plotted as a function of fractional error for Prasher's Aggregate Model, blue dots represent the upper limit and red dots represent the lower limit.
Similar comparison was done for aggregation model proposed by Prasher [31]. It is based on the assumption that the agglomeration of nanoparticles leads to formation of clusters which lead to better thermal conduction and lower the conduction resistance. The results were compared for a maximum and minimum thermal conductivity value based on the maximum and minimum aggregation states. The comparison has been shown in Figure 33 and Figure 34. The blue dots display the thermal conductivity at lower aggregation state (hence lower limit) and the red dots represent the thermal conductivity at higher aggregation state (hence upper limit). When used with a range of values for the aggregation parameter, this model is effective at bounding the data at almost all concentration values. Almost all calculated enhancement values lie within 10% of the corresponding measured values. Most of the results can be predicted correctly at lower and intermediate aggregation states. Figure 34 shows that almost 90% of the experimental results were predicted within 10% of the measured value at lower aggregation states while at higher aggregation states the thermal conductivity was over predicted.

The comparison of results of the INPBE exercise with different theoretical models was done to understand the underlying phenomenon responsible for thermal conductivity enhancement. To summarize following trends were observed:

- Maxwell’s static model reproduces the thermal conductivity data obtained during INPBE exercise very well. Almost 90% results are within 10% accuracy.
- Prasher’s aggregate model predictions bound the measured data with a maximum and minimum value for thermal conductivity obtained by assuming minimum and maximum possible aggregation.
- Dynamic models tend to overestimate enhancement, particularly at high concentration and in non-aqueous nanofluids (Prasher’s Model)
- No nanofluid sample seems to have abnormally high thermal conductivity (defined as a very significant deviation from Maxwell’s static model)
- Sensitivity analysis suggests that the uncertainties in assumed parameters produces no significant change in the obtained results from theoretical models
3. Analysis of Anomalous Convective Heat Transfer Enhancement in Nanofluids

3.1. Methodology

As discussed in Section 1, nanofluids have different heat transfer coefficients than the corresponding basefluids because their thermo-physical properties such as thermal conductivity, specific heat, viscosity and density are different as compared to the basefluid. The difference in their properties arises primarily due to the addition of nanoparticles to the basefluids. These nanoparticles have higher thermal conductivity and hence tend to enhance the heat transfer characteristics of the nanofluid as compared to the corresponding basefluid.

a) Collection of Data

There have been several experimental studies which suggest that enhancement in heat transfer coefficient for nanofluids is more than what can be predicted by the classical theoretical correlations for laminar and turbulent flows. The aim of the research was to investigate if such claims were correct by evaluating the heat transfer coefficients using equation 6 for laminar flow and equation 5 for turbulent flow regimes. First, research was conducted to compile the published data on nanofluid heat transfer. The scope was determined by narrowing the focus of the investigation to one type or a class of nanofluids for which there is published data about convective heat transfer coefficients, as well as information about particle size, particle concentration, and experimentation method. More than 40 published experimental studies on nanofluid heat transfer coefficients [54-103] were found during this research exercise. Out of these, based on availability of data and claims of anomalous enhancement 12 studies [93-103] were shortlisted for analysis of claims of anomalous enhancement. 8 studies [93-100] on laminar flow report enhancement beyond theoretical prediction by equation 6 while 4 studies [100-103] on turbulent flow report enhancement beyond theoretical estimation by equation 5. Experimental measurement data and reported data such as Reynolds number, volumetric flow rate, heat transfer coefficient, viscosity, thermal
conductivity, density, specific heat and other useful datum if provided was extracted and used if necessary for the comparison and estimation of theoretical value of heat transfer correlation. If necessary the data was extracted from the figures reported in the published studies using a ruler.

b) Calculation of theoretical heat transfer coefficient

For heat transfer coefficient, the database on nanofluid convective heat transfer was critically evaluated with respect to the possibility of abnormal enhancement above the values predicted by existing heat transfer correlations, when all properties are included in the calculations. To complete this thesis objective, the relevant experimental data was used from [93-103].

The Dittus-Boelter correlation for convective heat transfer in fully-developed turbulent flow given by equation 5 was used to evaluate the HTC for turbulent flow experimental studies [100, 103]:

\[ Nu = 0.023 \text{Re}^{0.8} \text{Pr}^{0.4} \]

\[ h = 0.023 \text{Re}^{0.8} \frac{\epsilon^{0.4} k^{0.6} \mu^{0.4}}{D} = 0.023 \frac{\epsilon^{0.4} k^{0.6} \rho^{0.4} V^{0.8}}{\mu^{0.4} D^{0.2}} \quad (12) \]

where, \( h, \rho, \mu \) and \( k \) are the convective heat transfer coefficient, density, viscosity and thermal conductivity of the nanofluid respectively. \( D \) and \( V \) are the diameter of the pipe and the velocity of the nanofluid respectively. Equation 12 is valid for \( 0.7 < Pr < 120, \text{Re} > 10,000 \) and \( L/D > 10 \).

Some turbulent flow studies [101, 102] compared their data to the Gnielinski correlation for turbulent flow as given by equation 13

\[ Nu_D = \frac{f/8(Re_D - 1000)Pr}{1 + 12.7 \left( \frac{f}{8} \right)^{1/2} (Pr^{2/3} - 1)} \quad (13) \]

where \( f = (0.79 \ln Re_D - 1.64)^{-2} \quad (14) \)

Equation 13 is valid for \( 5000 < \text{Re} < 5 \times 10^6 \) and \( 0.5 < Pr < 2000 \).
The correlation given by equation 15 for laminar flow reproduces the complex analytical solution for local Nusselt number to within 1%. Equation 15 was used to estimate local heat transfer coefficient for the laminar flow studies:

$$Nu_x = \begin{cases} 
1.302(x^+)^{-1/3} - 1, & x^+ \leq 0.0005 \\
1.302(x^+)^{-1/3} - 0.5, & 0.0005 \leq x^+ \leq 0.0015 \\
4.364 + 8.68(10^3x^+)^{-0.506}e^{-(41x^+)} & x^+ \geq 0.0015
\end{cases} \tag{15}$$

where, $Nu_x = \frac{h(x)D}{k}$ and the dimensionless distance, $x^+ = \frac{(x/D)}{RePr}$. Equation 15 is valid for $Re < 2300$.

Some of the laminar flow studies reported average heat transfer coefficient over the entire heat transfer region. This was predicted using equation 16 given by Shah [107] for average Nusselt number:

$$Nu_m = \begin{cases} 
1.953 x^*-1/3 - 1 & \text{for } x^* \leq 0.03 \\
4.364 + 0.0722 & \text{for } x^* > 0.03
\end{cases} \tag{16}$$

where $x^* = \frac{(L/D)}{RePr}$. L and D are the length and diameter of the test section. Equation 16 is valid for $Re < 2300$.

Heris et al [93] measured laminar heat transfer coefficient at constant wall temperature boundary condition and reported the average heat transfer coefficient. Equations 17 and 18 given by the Graetz and Nusselt were used for the theoretical prediction of heat transfer coefficient:

$$\theta_m = \frac{T_w - T_m}{T_w - T_c} = 8 \sum_{n=0}^\infty \frac{G_n}{\lambda_n^2} \exp(-2 \lambda_n^2 x^*) \tag{17}$$

$$Nu_m,\theta = -\frac{\ln \theta_m}{4x^*} \tag{18}$$

where $x^* = \frac{(L/D)}{RePr}$ and $\lambda_n$ represents the eigenvalues of the eigenfunctions of the analytical solution.

Values of $\lambda_n$ and $G_n$ can be found from Table 5.3 on page 5.10 of the heat transfer handbook [106].
Because of the contradictory claims put forth by different studies of nanofluids [93-103] about the prediction of heat transfer in nanofluids by conventional heat transfer correlations, published results reporting anomalous heat transfer enhancement were re-evaluated with updated nanofluid properties to correctly include the effect of addition of nanoparticles on 4 main nanofluid properties: thermal conductivity ($k$), viscosity ($\mu$), specific heat ($C_p$), density ($\rho$). Nusselt number was predicted by the correlations using the Reynolds and Prandtl numbers which were either reported or calculated with the measured temperature- and loading-dependent properties. Finally heat transfer coefficient was calculated using the loading and temperature dependent thermal conductivity which was either reported in the published results or recalculated using the Nan et al [105] model which was found to be predicting the thermal conductivity within 10% for the INPBE study.

Shortlisted set of publications [93-103], were investigated. The data that were used to conduct the study included nanoparticle and base fluid composition, particle size, particle concentration in the nanofluid, any other pertinent properties of the fluid and its constituents, such as surfactants or additives uses, and the results of the experiments performed to determine the convective heat transfer coefficient. Data were extracted from published figures if the data was not presented in a tabular form.

Appropriate correlation for heat transfer coefficient was evaluated using the Reynolds and Prandtl number, which were either reported or calculated from measured temperature- and loading-dependent thermal conductivities. These calculations were performed using an excel spreadsheet with extracted data, nanoparticle properties and basefluid properties as inputs. Once the expected theoretical heat transfer coefficient was evaluated using the corrected properties of nanofluids, it was compared to the experimentally measured heat transfer coefficient to determine whether there is any enhancement of heat transfer in the nanofluids beyond what can be predicted by the established correlations.
c) **Definition of Anomalous Enhancement**

The data was plotted with heat transfer coefficient on the y axis and either Re or x/D on the x axis. The choice of x-axis was based on the availability of data and experimental measurement technique used by the researchers. Error bars associated with the uncertainty in the experimental measurement were plotted along the y axis. Similar error bars associated with uncertainty in the theoretical estimation of the heat transfer coefficient were also plotted along the y axis. Hence each experimental measurement had a corresponding theoretical estimation both with their error bars. If the error bars of experimental values were clearly separated from the error bars of the corresponding theoretical estimation with the experimental value being higher, the enhancement was called as *anomalous* since it was beyond experimental measurement error and theoretical estimation error even after accounting for the properties of nanofluids in the theoretical correlations.

### 3.2. Error Analysis

Error estimation is a vital part of any analytical study as it gives an idea about the confidence in the results of the analysis. Error analysis was performed for each of the studies analyzed in this work. Error could be present either in the experimental results reported in the papers or in the theoretical estimation of the heat transfer coefficient due to uncertainty in the prediction by the theoretical model or models/experimental techniques used to measure nanofluid properties.

#### 3.2.1. Evaluation of Uncertainty in Theoretical Prediction

- Uncertainty in the nanoparticle properties namely $C_p$, $k_p$, and $\rho_p$ were conservatively assumed to be 10% in each of the studies analyzed. Although the nanoparticle properties are easily available through online databases and materials safety datasheets but there is an uncertainty associated with the change in physical properties of nanoparticles once they are dispersed in basefluid. Interactions of nanoparticles with basefluid molecules such as hydroxylation [109,110] and interaction with surfactants [111] could affect their physical properties.
• Nanofluid properties were either calculated using theoretical models or were taken from the studies if they were measured experimentally with reported experimental error. In case the nanofluid properties were predicted using theoretical models, uncertainties in the nanofluid properties were calculated by standard propagation of uncertainties in physical properties of nanoparticle through the theoretical models and model uncertainty was also taken into account. The model uncertainty (aleatory uncertainty) and input uncertainty (epistemic uncertainty) were treated independent of each other. If the nanofluid properties were reported as experimentally measured, the reported uncertainty in the experimental method or the best error estimate for the experimental technique used from the available literature was used.

• Finally the heat transfer coefficient was estimated using the nanofluid properties uncertainty and the heat transfer coefficient was calculated by propagating the uncertainties in the nanofluid properties through the theoretical model used for HTC prediction. Theoretical error is estimated by the simple error propagation as discussed in Appendix B to obtain the HTC within ±σ of the actual value.

3.2.2. Uncertainty in Experimentally Measured Values

Uncertainties in the experimental values were generally reported in the studies analyzed. If the experimental uncertainty was not reported, a generic uncertainty of 5% was attached to the experimental heat transfer coefficient. Typical uncertainty in heat transfer coefficient measurement experiments was found to be 3-5%. Table V listed below shows what uncertainties were imposed on different properties, experimental techniques and theoretical models for each of the experimental studies analyzed in this work.
### Table VII Experimental studies analyzed for laminar flow convective heat transfer coefficient

<table>
<thead>
<tr>
<th>Paper Authors</th>
<th>Nanofluid Characterization</th>
<th>Particle Size and Shape</th>
<th>Nanoparticle Concentration</th>
<th>Reynolds Number/Flowrate Range</th>
<th>Boundary Condition</th>
<th>Properties Calculation/Measurement</th>
<th>Result/Conclusion</th>
</tr>
</thead>
<tbody>
<tr>
<td>Anoop et al [95]</td>
<td>Al₂O₃/water Nanofluid</td>
<td>45 nm and 150 nm Spherical</td>
<td>0.26 vol %, 0.52 vol %, 1.06 vol%, 1.61 vol %, 2.18 vol %</td>
<td>750-2200</td>
<td>Constant heat flux DC Power Supply</td>
<td>THW Ubchelohde viscometer Mixture formula Mixture formula</td>
<td>Anomalous enhancement observed with an increasing trend at higher concentration and higher Reynolds number</td>
</tr>
<tr>
<td>Ding et al [96]</td>
<td>CNT/water nanofluid</td>
<td>Aspect ratio &gt;100</td>
<td>0.10 wt%, 0.25 wt%, 0.50 wt%</td>
<td>800-1200</td>
<td>Constant heat flux DC Power Supply</td>
<td>KD-2 Bohlin CVO Rheometer Mixture formula Mixture formula</td>
<td>Very high enhancement observed in the developing region of the laminar flow regime. It is surely anomalous.</td>
</tr>
<tr>
<td>Authors</td>
<td>Nanofluid Type</td>
<td>Nanoparticle Size</td>
<td>Nanoparticle Concentration</td>
<td>HTC Measurement Method</td>
<td>HTC Calculation Method</td>
<td>Nanofluid Percentage</td>
<td>Notes</td>
</tr>
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</tr>
<tr>
<td>Heris et al [93]</td>
<td>Al₂O₃/water Nanofluid</td>
<td>20 nm Spherical</td>
<td>0.1, 1.0, 2.0, 2.5, 3.0 vol%</td>
<td>N/A</td>
<td>Constant Wall Temperature</td>
<td>N/A</td>
<td>Max well's equation was observed and the enhancement increased with increasing Peclet number and increasing concentration of nanoparticles.</td>
</tr>
<tr>
<td>Choi et al [98]</td>
<td>Al₂O₃/water Nanofluid</td>
<td>30±5 nm Spherical</td>
<td>0.01, 0.02, 0.03, 0.1, 0.2, 0.3 vol%</td>
<td>400-750 Flowrate controlled by HV77921-40 pump</td>
<td>Constant Heat flux AC Power Supply</td>
<td>N/A</td>
<td>THW, VM 10-A Viscometer, BX 300 DSC 204 F1 Anomalous enhancement was confirmed only at 0.3 vol% concentration.</td>
</tr>
<tr>
<td>Kurowska et al [98]</td>
<td>151 nm (0.15 vol%) Cu/Ethylene Glycol Nanofluid</td>
<td>Spherical</td>
<td>0.15, 0.25 vol%</td>
<td>30-60 Flowrate determined by flow meter (±0.01 l/hr)</td>
<td>Constant heat flux DC Power Supply</td>
<td>N/A</td>
<td>Brookfield DV II+ Viscometer, Mixture Formula Only vol fraction weighed. Anomalous enhancement seen only at the entrance point of the flow regime.</td>
</tr>
<tr>
<td>Prasher et al [97]</td>
<td>20 nm (SEM) Aggregate size (100-300)</td>
<td>Spherical</td>
<td>0.5, 0.75, 1.00 vol%</td>
<td>1 - 9 (ml/min) (Micro)</td>
<td>Constant heat flux DC Power</td>
<td>N/A</td>
<td>Mixture formula, Mixture formula Higher enhancement at higher concentration.</td>
</tr>
<tr>
<td>Author</td>
<td>Particle Size</td>
<td>Shape</td>
<td>Concentration</td>
<td>Flowrate</td>
<td>Heat Flux</td>
<td>DC Power Supply</td>
<td>THW</td>
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</tr>
<tr>
<td>Li et al [100]</td>
<td>&lt;100 nm</td>
<td>Spherical</td>
<td>0.3 vol %, 0.5 vol %, 0.8 vol %, 1.0 vol %, 1.2 vol %, 1.5 vol %, 2.0 vol %</td>
<td>850-2200</td>
<td>Constant heat flux</td>
<td>DC Power Supply</td>
<td>THW</td>
</tr>
<tr>
<td>Ding et al [94]</td>
<td>27-56 nm</td>
<td>Spherical</td>
<td>0.6 vol %, 1.00 vol %, 1.60 vol %</td>
<td>1050, 1600</td>
<td>Constant heat flux</td>
<td>DC Power Supply</td>
<td>KD-2</td>
</tr>
</tbody>
</table>
Table VIII Experimental studies analyzed for turbulent flow convective heat transfer coefficient

<table>
<thead>
<tr>
<th>Paper Authors</th>
<th>Nanofluid Characterization</th>
<th>Volumetric Concentration</th>
<th>Reynolds Number Range</th>
<th>Boundary Condition</th>
<th>Properties Calculation/Measurement</th>
<th>Result/Conclusion</th>
</tr>
</thead>
<tbody>
<tr>
<td>Xuan and Li [100]</td>
<td>Cu/water nanofluid</td>
<td>N/A</td>
<td>0.3%, 0.5%, 0.8%, 1.0%, 1.2%, 1.5%, 2.0%</td>
<td>10,000-25,000</td>
<td>Constant heat flux</td>
<td>THW, NXE-1 viscometer, Mixture formula</td>
</tr>
<tr>
<td>Duangthong and Wongwises [101]</td>
<td>TiO2/water nanofluid</td>
<td>N/A</td>
<td>0.2%</td>
<td>4000-18000</td>
<td>Flowrate determined by electronic balance</td>
<td>Yu and Choi's Model(^5), Einstein formula</td>
</tr>
</tbody>
</table>
**Pak and Cho [103]**

<table>
<thead>
<tr>
<th>System</th>
<th>Nanoparticle Size</th>
<th>Aspect Ratio</th>
<th>Al₂O₃ Concentration</th>
<th>Heat Flux</th>
<th>Method</th>
<th>Measurement</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>TiO₂/water</td>
<td>27 nm (TiO₂)</td>
<td>~1</td>
<td>1.34%, 2.78%</td>
<td>14000-60000</td>
<td></td>
<td>Masuda et al 6</td>
<td>Anomalous enhancement was seen both for titania and alumina nanofluid beyond 1 vol% conc. The enhancement increased at higher nanoparticle concentration and also at higher Reynolds number.</td>
</tr>
<tr>
<td></td>
<td>13 nm (Al₂O₃)</td>
<td>~1</td>
<td>TiO₂ (0.99%, 2.04%, 3.16%)</td>
<td></td>
<td>Brook field cone and plate viscometer</td>
<td></td>
<td>Anomalous enhancement is observed only under conditions of viscosity of nanofluid estimated at room temperature. As discussed in Section 3.4, temperature variation of viscosity can have a significant effect on the analysis and the experimental and theoretical curves shift closer to each other if viscosity is estimated at higher temperature which can be the source of error in the analysis due to unknown experimental temperatures.</td>
</tr>
</tbody>
</table>

**Yu et al [102]**

<table>
<thead>
<tr>
<th>System</th>
<th>Nanoparticle Size</th>
<th>Aspect Ratio</th>
<th>Concentration</th>
<th>Heat Flux</th>
<th>Method</th>
<th>Measurement</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiC/water nanofluid</td>
<td>~170 nm (DLS and Small angle X-ray scattering)</td>
<td>Disks or platelets</td>
<td>3.7%</td>
<td>3,000-12,000</td>
<td>Constant heat flux</td>
<td>THW DV-II+Proviscometer</td>
<td>Anomalous enhancement observed which increases with higher Reynolds number</td>
</tr>
<tr>
<td>Paper Authors</td>
<td>Nanofluid Property Estimation Uncertainty</td>
<td>Mass Flowrate Uncertainty $\bar{m}$ (kg/s)</td>
<td>Experimental HTC Uncertainty</td>
<td></td>
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<tr>
<td></td>
<td>Thermal Conductivity $k$ (W/m-K)</td>
<td>Viscosity $\mu$ (Pa.s)</td>
<td>Density $\rho$ (kg/m$^3$)</td>
<td>Specific Heat $C_p$ (J/kg-K)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Anoop et al [95]</td>
<td>THW (±2%)</td>
<td>a</td>
<td>Mixture Formula</td>
<td>Mixture Formula</td>
<td>±5% (assumed)</td>
<td>±5% (assumed)</td>
<td></td>
</tr>
<tr>
<td>Ding et al [96]</td>
<td>KD2 (±3%)</td>
<td>a</td>
<td>Mixture Formula</td>
<td>Mixture Formula</td>
<td>±4.6% (reported)</td>
<td>±5% (assumed)</td>
<td></td>
</tr>
<tr>
<td>Heris et al [93]</td>
<td>Maxwell’s model (±10% assumed from INPBE)</td>
<td>a</td>
<td>Mixture Formula</td>
<td>Mixture Formula</td>
<td>±5% (assumed)</td>
<td>±5% (assumed)</td>
<td></td>
</tr>
<tr>
<td>Choi et al [98]</td>
<td>THW (±5%, assumed from INPBE)</td>
<td>a</td>
<td>Mixture Formula</td>
<td>Mixture Formula</td>
<td>±5% (assumed)</td>
<td>±3%</td>
<td></td>
</tr>
<tr>
<td>Kurowska et al [99]</td>
<td>Maxwell’s model (±10% assumed from INPBE)</td>
<td>a</td>
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<td>Mixture Formula</td>
<td>±5% (assumed)</td>
<td>±5% (assumed)</td>
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<td>a</td>
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<td>±5% (assumed)</td>
<td>±5% (assumed)</td>
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<tr>
<td>Li et al [100]</td>
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<td>Mixture Formula</td>
<td>Mixture Formula</td>
<td>±1% (reported)</td>
<td>±4%</td>
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<tr>
<td>Ding et al [94]</td>
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<td>Li et al [100]</td>
<td>THW (±5%, assumed from INPBE)</td>
<td>NXE-1 viscometer (±5% measurement uncertainty due to volume fraction. ±10% uncertainty assumed due to temperature)</td>
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<td>±1% (reported)</td>
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<tr>
<td>Duangthongsuk and Wongwises [101]</td>
<td>Maxwell’s model (±10% assumed from INPBE)</td>
<td>Mean of MIT data and Einstein’s model (^b) (±4.32% model uncertainty, calculated by using the bounds as)</td>
<td>Mixture Formula</td>
<td>Mixture Formula</td>
<td>±5% (assumed)</td>
<td>±5% (assumed)</td>
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</table>
Laminar flow correlations require the calculation of Peclet number, \( Pe = \frac{\rho v D \Delta T}{k} \) only. Since \( Pe \) number does not account for viscosity, we do not need to estimate the viscosity of the medium to calculate the heat transfer coefficient using correlations.

Turbulent flow convective heat transfer studies did not report viscosity measurement procedure/technique. In such cases the viscosity bounds had to be estimated. Williams et al [55] at MIT performed experiments to measure viscosity of nanofluids. They devised a formula using their

---

| Pak and Cho [103] | Maxwell’s model (±10% assumed from INPBE) | Brookfield viscometer (±2%, measurement uncertainty, ±10% temperature uncertainty) | MIT data and Einstein’s model, ±10.00% temperature uncertainty | Mixture Formula | Mixture Formula | ±5% (assumed) | ±5% (assumed) |
| Pak and Cho [103] | Maxwell’s model (±10% assumed from INPBE) | Brookfield viscometer (±2%, measurement uncertainty, ±10% temperature uncertainty) | MIT data and Einstein’s model, ±10.00% temperature uncertainty | Mixture Formula | Mixture Formula | ±5% (assumed) | ±5% (assumed) |

Yu et al [102] | THW (±5%, assumed from INPBE) | DV II+ Viscometer (±5%) | Mixture Formula | Mixture Formula | ±1% (reported) | ±5% |

---

\[ Pe = \frac{\rho v D \Delta T}{k} \]
experimental data and found the viscosity to be considerably higher than reported by classical Einstein formula, \( \mu = \mu_w(1 + 2.5\phi) \). The bounds for viscosity were calculated with maximum estimated by the formulae reported in [55] and the minimum by the Einstein’s formula.

\[
\text{Williams et al [55] viscosity measurement (upper limit)}
\begin{align*}
\text{For alumina nanofluid: } \mu(\phi, T) &= \mu_w(T)e^{4.91\phi}\left(1 + 0.2092\phi\right) \\
\text{For zirconia nanofluid: } \mu(\phi, T) &= \mu_w(T)e^{11.19\phi}\left(1 + 0.1960\phi\right)
\end{align*}
\]

Hence the viscosity was estimated to be the mean of the maxima and the minima. The uncertainty was estimated by the deviation of the mean from the maxima and minima.

c. Uncertainties in density and specific heat of nanofluid were calculated using error propagation through the mixture formula assuming standard 10% uncertainties in nanoparticle properties (Although nanoparticle properties can be determined with a good accuracy there is still an uncertainty as their physical properties might be affected by the interaction with basefluid molecules. Phenomenon like hydration are very common and are reported to effect the physical properties of nanofluids).
3.3. Experimental Studies Analyzed

The studies analyzed basically included published experimental work on laminar and turbulent flow regimes. Table VII and Table VIII list the various published experimental works which had reported an anomalous enhancement of heat transfer coefficient when compared to the prediction by the theoretical correlations.

3.3.1. Laminar Flow

Table VII lists the published studies which report anomalous enhancement of convective heat transfer coefficient during laminar flow. Heat transfer coefficient data was extracted from the papers and was compared to the theoretical correlation relevant to the experimental conditions used in the measurement.

Anoop et al [95] did an experimental investigation on the convective heat transfer characteristics in the developing region of tube flow with constant heat flux with alumina-water nanofluids. Two particle sizes were used, one with average particle size of 45 nm and the other with 150 nm. It was observed that in the developing region, the nanofluid heat transfer coefficients show higher enhancement with respect to the HTC of water than in the developed region. A new correlation was proposed by Anoop et al to predict the heat transfer coefficient for the range of nanofluids used in their experiments. Experimental data on measured heat transfer coefficient and Reynolds number was extracted only for 150 nm nanoparticles. The theoretical value of heat transfer coefficient was predicted using equation 15. Uncertainties and methods of estimation of nanofluid properties for predicting theoretical HTC have been listed in Table IX.

Viscosity was measured using Ubbelohde viscometer. Since equation 15 uses the Peclet Number \((Pe)\) which is the product of Reynolds number \(\left(Re = \frac{\rho u D}{\mu}\right)\) and Prandtl number \(\left(Pr = \frac{\mu C_p}{k}\right)\), effect of error in viscosity measurement is cancelled. Since the Reynolds number was directly extracted from the paper, precision was taken to use the same value of viscosity for the prediction of Prandtl number as was used by the authors in the estimation of Reynolds number so that the effect of viscosity measurement was
cancelled. Error in estimation of thermal conductivity using THW was reported to be 2%. Error in density and specific heat was estimated by propagation of uncertainties through formulae. Figure 35 to Figure 38 show the variation of both experimental and theoretical HTC with Reynolds number at x/D=147 (~halfway in the tube). An error bar is associated with each experimental measurement as well as theoretical estimation. It can be clearly seen that the HTC there is a clear anomalous enhancement which is not predicted by the regular theoretical correlation given by equation 15. This anomalous enhancement becomes more prominent (>15%) at higher Reynolds number and increases with higher concentration of nanoparticles.

Figure 38 shows the HTC variation along the tube during the developing flow regime at a Reynolds number 1550. It can be seen that there is a higher deviation of experimental value from the theoretical estimation and the enhancement is anomalous as the error bars do not overlap.
Figure 35 Local HTC vs Re for 1 wt% alumina (x/D=147; 150 nm particles) Anoop et al

Figure 36 Local HTC vs Re for 4 wt% alumina (x/D=147; 150 nm particles) Anoop et al

Figure 37 Local HTC vs Re for 6 wt% alumina (x/D=147; 150 nm particles) Anoop et al

Figure 38 HTC vs (x/D) for 4 wt% alumina (150 nm nanoparticles) Anoop et al
Figure 39 Local HTC in the developing region for 0.6 vol% alumina (Re 1600) Ding et al

Figure 40 Local HTC in the developing region for 1 vol% alumina (Re1600) Ding et al

Figure 41 Local HTC in the developing region for 1.6 vol% alumina (Re 1600) Ding et al

Figure 42 Local HTC in the developing region for 0.6 vol% alumina (Re 1050) Ding et al

Figure 43 Local HTC in the developing region for 1.0 vol% alumina (Re 1050) Ding et al

Figure 44 Local HTC in the developing region for 1.6 vol% alumina (Re 1050) Ding et al
Ding et al [93] did an experimental work on the convective heat transfer of nanofluids, made of $\gamma$-Al$_2$O$_3$ nanoparticles and de-ionized water, flowing through a copper tube in the laminar flow regime. The results showed considerable enhancement of convective heat transfer using the nanofluids. The enhancement was particularly significant in the entrance region, and was much higher than that solely due to the enhancement on thermal conductivity. It was also shown that the classical Shah [107] equation failed to predict the heat transfer behavior of nanofluids. Experimental data on heat transfer coefficient was extracted for the 0.6 vol%, 1.0 vol% and 1.6 vol% alumina nanofluids for Reynolds number of 1050 and 1600. Viscosity was estimated using Einstein model given by equation 19

$$\mu = \mu_f (1 + 2.5\phi)$$  \hspace{1cm} (19)

where $\mu_f$ is the viscosity of the basefluid and $\phi$ is the volume fraction of nanoparticles in the nanofluid. The calculated viscosity was used to estimate the Prandtl number which was used for estimation of theoretical HTC. Thermal conductivity was measured using KD2 instrument and was extracted from the paper. The uncertainty in the measured thermal conductivity was reported to be 3%. Other uncertainties were propagated using simple error propagation. Figure 39 to Figure 44 represent the comparison of theoretical predictions obtained from equation 15 with the experimental data extracted. It can be clearly observed that the heat transfer coefficient measured during experiments is considerably higher than the theoretical estimation after accounting for errors in theoretical estimation and experimental measurements. There is a clear anomalous enhancement and the deviation is more prevalent in the entrance region. Uncertainties and methods of estimation of nanofluid properties for predicting theoretical HTC have been listed in Table IX.
Figure 45 Average experimental HTC and average theoretical HTC plotted as a function of Peclet number for 0.2 vol% alumina nanofluid (Heris et al).

Figure 46 Average experimental HTC and average theoretical HTC plotted as a function of Peclet number for 1.0 vol% alumina nanofluid (Heris et al).

Figure 47 Average experimental HTC and average theoretical HTC plotted as a function of Peclet number for 2.0 vol% alumina nanofluid (Heris et al).

Figure 48 Average experimental HTC and average theoretical HTC plotted as a function of Peclet number for 2.5 vol% alumina nanofluid (Heris et al).

Figure 49 Average experimental HTC and average theoretical HTC plotted as a function of Peclet number for 3.0 vol% alumina nanofluid (Heris et al).
Heris et al [97] investigated nanofluids containing CuO and Al₂O₃ oxide nanoparticles in water as base fluid. The laminar flow convective heat transfer through circular tube with constant wall temperature boundary condition was examined. The experimental results emphasize that the single phase correlation with nanofluids properties (Homogeneous Model) is not able to predict heat transfer coefficient enhancement of nanofluids. Experimental data was extracted for 0.2 vol%, 1.0 vol%, 2.0 vol%, 2.5 vol% and 3.0 vol% Al₂O₃ nanofluid. Since constant wall temperature boundary condition was used during the experiments, equation 17 was used to predict the theoretical heat transfer coefficient. Peclet number was reported and hence there was no issue of viscosity measurement uncertainty. Thermal conductivity was estimated using equation 1 for effective medium theory. An uncertainty of 10% was attached to the model for prediction of thermal conductivity as was found during the INPBE exercise. The comparison of the theoretical HTC result with experimental values for different concentration has been shown in Figure 45 to Figure 49. HTC is plotted as a function of Peclet number. It can be clearly observed that there is a higher deviation of experimental values from the theoretical prediction at higher Peclet number. High Peclet number is equivalent to higher Reynolds number for a given concentration and hence the trend of higher deviation might be attributed to a more chaotic movement of nanoparticles at higher Reynolds number enhancing the heat transfer rate. A more detailed analysis needs to be done to understand the mechanism of this enhancement.

Choi et al [99] measured convective heat transfer coefficient of water-based Al₂O₃ nanofluids flowing through a uniformly heated circular tube in the fully developed laminar flow regime. The paper reports that the convective heat transfer coefficient of the nanofluids increases by up to 8% at a concentration of 0.3 vol% compared with that of pure water and this enhancement cannot be predicted by the Shah equation. Experimental HTC data was extracted for 0.1 vol%, 0.2 vol% and 0.3 vol% alumina nanofluid. Equation 15 was used to predict the experimental HTC after the estimation of nanofluid properties as per the methods listed in Table IX. Viscosity was measured using VM-10A viscometer while the thermal
conductivity was measured using transient hot wire method. Measured viscosity was used to estimate the Prandtl number. Error propagation was done to get the uncertainty in the value of

![Graph](image1.png)

Figure 50 Local theoretical HTC and experimental HTC plotted as a function of x/D in the developing flow region (Re =700, 0.1 vol% alumina) (Choi et al)

![Graph](image2.png)

Figure 51 Local theoretical HTC and experimental HTC plotted as a function of x/D in the developing flow region (Re =700, 0.2 vol% alumina) (Choi et al)

![Graph](image3.png)

Figure 52 Local theoretical HTC and experimental HTC plotted as a function of x/D in the developing flow region (Re =700, 0.3 vol% alumina) (Choi et al)

the HTC predicted by the theory. Uncertainty associated with each parameter is listed in Figure 50 to Figure 52 show the data for experimental and theoretical HTC after analysis. Although the experimentally measured HTC is consistently higher than the theoretically predicted HTC, the value is within the bounds of experimental error and theoretical estimation error except for 0.3 vol% concentration nanofluid. In other words, the enhancement cannot be really confirmed to be anomalous at concentrations lower than 0.3 vol% because of the uncertainty in the values of experimental and the theoretical HTC.
Kurowska et al [98] investigated convective heat transfer in nanofluid based on Cu (approx. 0.15 vol % and 0.25 vol %) nanoparticles at constant heat flux conditions. A 30% increase in average heat transfer coefficient was reported against the results obtained for a pure host liquid (ethylene glycol). Even more significant increase was reported in the entrance region. Experimental HTC data was extracted for 0.15 vol% Cu- nanofluid. Temperature dependent nanofluid viscosity was estimated using empirical formula of type $\eta = aT^2 + bT + c$ as given by Eastman et al [108]. The constants a, b and c were determined for both the nanofluids using the Brookfield DV II+ viscometer. The temperature for the estimation of nanofluid properties was 298 K. Viscosity thus estimated was used in the estimation of Pr number to avoid viscosity measurement errors. Effective medium theory was used to estimate the thermal conductivity of nanofluid with 10% accuracy as reported in the INPBE study. Figure 53 and Figure 54 show the comparison of experimental measurements reported with the theoretical estimation. Figure 53 shows that there is an anomalous enhancement at the beginning of the entrance region but the enhancement is very nearly predicted by the theory once the flow starts to develop. Figure 54 shows that average HTC generally increases with increasing Reynolds number but anomalous enhancement could not be confirmed. The wavy behavior of the experimental curve is reported to experimental errors.
Prasher et al [98] report the heat transfer coefficient in both developing and fully-developed regions by using water based alumina nanofluids. Experimental test section consists of a single 1.02-mm-diameter stainless steel tube which is electrically heated to provide a constant wall heat flux. Experiments were performed with 0.5 vol%, 0.75 vol% and 1.0 vol% Al₂O₃ nanofluids for 3 different flow-rates 1 ml/min, 5 ml/min and 9 ml/min. Data was extracted from the paper and the theoretical estimation was done using equation 15. Thermal conductivity was predicted the effective medium theory with an uncertainty of 10% which was taken into account during error analysis. Viscosity was estimated using Einstein’s model. This viscosity was used for estimation of both Reynolds and Prandtl number. Hence error in viscosity doesn’t matter since it cancels out in the Peclet number (Pe=Re.Pr). After analysis of error, the results obtained are shown in Figure 55 to Figure 63. Concentration of nanoparticles in these nanofluids is very low and no anomalous enhancement seems obvious. A trend of higher deviation from theoretical values at higher concentration and higher flowrate (Reynolds number) is seen. Although results of the experimental measurements are consistently above the theoretically estimated value, they are within the bounds of theoretical and experimental uncertainty and hence any anomalous enhancement couldn’t be confirmed.

![Figure 55](image1.png)  
**Figure 55** Local experimental and theoretical HTC (0.5 vol% alumina, 9ml/min)  
Prasher et al

![Figure 56](image2.png)  
**Figure 56** Local experimental and theoretical HTC (0.5 vol% alumina, 5ml/min)  
Prasher et al
Figure 57 Local experimental and theoretical HTC (0.5 vol% alumina, 1 ml/min)  
Prasher et al

Figure 58 Local experimental and theoretical HTC (0.75 vol% alumina, 9 ml/min)  
Prasher et al

Figure 59 Local experimental and theoretical HTC (0.75 vol% alumina, 5 ml/min)  
Prasher et al

Figure 60 Local experimental and theoretical HTC (0.75 vol% alumina, 1 ml/min)  
Prasher et al

Figure 61 Local experimental and theoretical HTC (1.0 vol% alumina, 9 ml/min)  
Prasher et al

Figure 62 Local experimental and theoretical HTC (1.0 vol% alumina, 5 ml/min)  
Prasher et al
Figure 63 Local experimental and theoretical HTC (1.0 vol% alumina, 1 ml/min)

Prasher et al.

0.3 vol % Cu

Figure 64 Average theoretical and experimental HTC vs Reynolds number (0.3 vol% Cu) (Li et al)

0.5 vol% Cu

Figure 65 Average theoretical and experimental HTC vs Reynolds number (0.5 vol% Cu) (Li et al)

0.8 vol% Cu

Figure 66 Average theoretical and experimental HTC vs Reynolds number (0.8 vol% Cu) (Li et al)

1.0 vol% Cu

Figure 67 Average theoretical and experimental HTC vs Reynolds number (1.0 vol% Cu) (Li et al)
Li et al [94] investigated convective heat transfer and flow characteristics of Cu-water nanofluid in a tube for the laminar and turbulent flow. They discussed the effects of such factors as the volume fraction of suspended nanoparticles and the Reynolds number on the heat transfer and flow characteristics in detail. They report that their experimental results show the suspended nanoparticles remarkably increase the convective heat transfer coefficient of the base fluid and showed that the friction factor of the sample nanofluid with the low volume fraction of nanoparticles is almost not changed. Compared with the base fluid, for example, they reported that the convective heat transfer coefficient increased about 60% for the nanofluid with 2.0 vol% Cu nanoparticles at the same Reynolds number. The different concentrations of
Cu-water nanofluid used for this study were 0.2 vol%, 0.5 vol%, 0.8 vol%, 1 vol%, 1.2 vol%, 1.5 vol% and 2.0 vol%. The experimental data was extracted from the paper and theoretical estimation of HTC was made using equation 13 using nanofluid properties estimated as per methods given in Table IX. Thermal conductivity was measured using transient hot wire method. Viscosity was measured using a NXE-1 viscometer. Thermal conductivity and viscosity were both measured with 5% accuracy. Error analysis was performed and the results are shown in Figures 64 to 70. It can be clearly seen that the experimentally measured values for the HTC are way higher than the theoretically estimated HTC. The enhancement seen in these cases is certainly anomalous as it is way beyond the experimental and theoretical estimation uncertainty. Moreover the enhancement is higher at a higher concentration and a higher Reynolds number, a trend which has been consistently observed across most of the data analyzed. Ding et al [96] investigated the heat transfer behavior of aqueous suspensions of multi-walled carbon nanotubes (CNT nanofluids) flowing through a horizontal tube. They reported that significant enhancement of the convective heat transfer is observed and the enhancement depends on the flow conditions (Reynolds number, Re), CNT concentration and the pH. Two nanofluids with 0.1 wt% and 0.5 wt% concentration were tested. Thermal conductivity was measured with KD2 property meter with 3% accuracy while viscosity was measured using CVO rheometer. Measured viscosity and thermal
conductivity was used for prediction of Prandtl number. After HTC data extraction and comparison with theoretically predicted HTC the plots have been shown in Figure 71 and Figure 72.

3.3.2. Turbulent Flow

Four experimental studies were analyzed for turbulent flow to check if the claims of the anomalous enhancement in HTC were valid under the experimental and theoretical uncertainty limits.

Xuan and Li [94] investigated convective heat transfer and flow features of Cu-water nanofluid in a tube. Both the convective heat transfer coefficient and friction factor of the sample alumina nanofluids for the turbulent flow were measured, respectively and new type of convective heat transfer correlation was proposed to correlate experimental data of heat transfer for nanofluids. The alumina nanofluids having concentrations 0.3 vol%, 0.5 vol%, 0.8 vol%, 1.0 vol%, 1.2 vol%, 1.5 vol%, and 2.0 vol% were used during this study. Thermal conductivity was measured using transient hot wire method. Viscosity was measured using a NXE-1 viscometer. Thermal conductivity and viscosity were both measured with 5% accuracy. The experimental data was extracted and compared to theoretical values with uncertainty attached to each value. The uncertainty in theoretical estimation was determined by error propagation and the uncertainties in the different parameters have been listed in Table IX. The results are shown in Figure 73 to Figure 79. At lower concentration the experimentally measured values are very close to the ones predicted by theory. The plots suggest that the experimental enhancement is always above the theoretically predicted value but it can be clearly distinguished only at a higher concentration of nanoparticles (>1 vol%) and also the enhancement deviates from the theoretical value at higher Reynolds number.
Figure 73 Average theoretical and experimental HTC vs Reynolds number (Li et al)

Figure 74 Average theoretical and experimental HTC vs Reynolds number (Li et al)

Figure 75 Average theoretical and experimental HTC vs Reynolds number (Li et al)

Figure 76 Average theoretical and experimental HTC vs Reynolds number (Li et al)

Figure 77 Average theoretical and experimental HTC vs Reynolds number (Li et al)

Figure 78 Average theoretical and experimental HTC vs Reynolds number (Li et al)
Duangthongsuk and Wongwises [101] studied the forced convective heat transfer and flow characteristics of a nanofluid consisting of water and 0.2 vol% TiO$_2$ nanoparticles. The heat transfer coefficient and friction factor of the TiO$_2$–water nanofluid flowing in a horizontal tube-in-tube counter flow heat exchanger under turbulent flow conditions were investigated. The results reported in this study show that the convective heat transfer coefficient of nanofluid is slightly higher than that of the base liquid by about
6–11%. The heat transfer coefficient of the nanofluid increases with an increase in the mass flow rate of the hot water and nanofluid, and increases with a decrease in the nanofluid temperature, and the temperature of the heating fluid has no significant effect on the heat transfer coefficient of the nanofluid. They also reported that the Gnielinski equation failed to predict the heat transfer coefficient of the nanofluid. Figure 80 shows the experimental data and theoretically predicted data on the HTC vs Re plot. It is clear from the plot that the enhancement is really anomalous as it is not predicted by the theory within the bounds of experimental/theoretical error.

![Figure 81 Experimental and theoretical HTC vs Reynolds number (Pak and Cho)](image1)

![Figure 82 Experimental and theoretical HTC vs Reynolds number (Pak and Cho)](image2)

![Figure 83 Experimental and theoretical HTC vs Reynolds number (Pak and Cho)](image3)

![Figure 84 Experimental and theoretical HTC vs Reynolds number (Pak and Cho)](image4)
Pak and Cho [103] investigated turbulent heat transfer behaviors of dispersed fluids (ultrafine metallic oxide particles suspended in water) in a circular pipe. Two different metallic oxide particles, $\gamma$-alumina ($\text{Al}_2\text{O}_3$) and titanium dioxide ($\text{TiO}_2$) with mean diameters of 13 and 27 nm, respectively, were used as suspended particles. The Reynolds and Prandtl numbers varied in the ranges $10^4$-$10^5$ and 6.5-12.3, respectively. Nanofluids investigated included 0.99 vol%, 2.04 vol%, 3.16 vol% titania nanofluids and 1.34 vol%, 2.78 vol% alumina nanofluids. Relevant experimental data was extracted from the paper and theoretical estimation of HTC was made using the Dittus Boelter correlation as given by equation 5. The data has been plotted in Figure 81 to Figure 85. It can be seen from the plots that there is definitely an anomalous enhancement observed in case of 2.78 vol% alumina and 3.16 vol% titania. At lower concentrations, however the enhancement couldn’t be confirmed under the limits of experimental measurement uncertainty and theoretical estimation error. However it is again noted that the experimentally measured value has a higher deviation at higher Reynolds number.

Yu et al [102] performed experiments with a water-based nanofluid containing 170-nm silicon carbide particles at a 3.7% volume concentration and having potential commercial viability. Heat transfer
coefficients for the nanofluid were presented for Reynolds numbers ranging from 3300 to 13,000 and are compared to the base fluid water. Relevant experimental data was extracted. Final plot of HTC vs Re is shown in Figure 86. There appears to be a significant deviation of experimental HTC from theoretical beyond the uncertainties in the experimental measurement and theoretical estimation. The enhancement shows higher deviation at higher Reynolds number, a trend observed in most of the studies analyzed. However this enhancement is observed when we have evaluated the viscosity at room temperature. As explained in Section 3.4 temperature variation of viscosity could significantly affect the analysis. As we see in the following section the experimental and theoretical curve for HTC move closer at higher temperature.

![3.7 vol% SiC nanofluid](image)

Figure 86 Experimental and theoretical HTC vs Reynolds number (Yu et al)
3.4. Interpretation of Results

3.4.1. Turbulent Flow

During this research exercise, 4 turbulent flow experimental studies [100-103] claiming an anomalous enhancement in the heat transfer coefficient beyond the prediction by theoretical models were analyzed. On analysis it was observed that the experimental heat transfer coefficient measured at the reported Reynolds number was higher than the theoretical prediction by Dittus Boelter correlation or Gnielinski correlation. Although this seems anomalous even after accounting for theoretical and experimental errors, one of the things which still remain unknown is the fluid temperature during the experiments. Due to lack of information on temperature of the experiments the properties of nanofluids were evaluated at room temperature. However, the correlations require the properties of the nanofluid at an average temperature of the heated section. This could affect the estimation of the heat transfer coefficient from the correlation.

It was shown in INPBE that the temperature effect on the thermal conductivity of nanofluid is not very significant in the temperature range 25 C-35 C and density and specific heat are also not strong functions of temperature. Hence the most dominant effect of temperature is on the viscosity of nanofluid. The

![Variation of viscosity of water with temperature](image)

*Figure 87 Variation of viscosity of water with temperature*
temperature effect of viscosity on nanofluid is captured by the change in viscosity of the basefluid with temperature as shown by Williams et al [55] for alumina and zirconia nanofluids. The general variation of nanofluid viscosity $\mu_{nf}$ is given by equation (10)

$$\mu_{nf}(\phi, T) = \mu_{bf}(T)f(\phi)$$

(10)

---

Table X Water Viscosity Data

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<tr>
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<tr>
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<td>0.2822</td>
<td>31%</td>
<td>1.59</td>
<td>59%</td>
<td>3.188873</td>
</tr>
</tbody>
</table>
where $\mu_b(T)$ is the temperature dependent viscosity of basefluid and $f(\phi)$ is an arbitrary functional dependence on volumetric fraction, $\phi$ of nanoparticles which is determined by experiments.

Figure 88 Effect of nanofluid temperature on theoretical and experimental HTC

Figure 89 Effect of nanofluid temperature on theoretical and experimental HTC
The studies analyzed during this research exercise for turbulent flow heat transfer coefficient had used water based nanofluids. Hence it was worthwhile to look at the temperature variation of viscosity of water. The temperature variation of viscosity of water has been shown in Figure 87.

As shown in Table X, at 55 C the viscosity of water drops almost to 56% of the room temperature value. Hence temperature variation of viscosity can significantly affect the value of the heat transfer coefficient prediction by the correlations. Table X indicates that the value of Nusselt number for turbulent flow estimated by Dittus Boelter correlation increases by almost 26% as compared to room temperature value, if the fluid temperature of experiment would be 55 C. Hence the theoretical prediction of heat transfer coefficient will be 26% higher for a given Reynolds number at 55 C. Hence there is a considerable amount of error induced in the prediction of heat transfer coefficient due to estimation of viscosity at room temperature (25 C). In general a temperature higher than room temperature (which is obvious for experiments) would cause the theoretical prediction to move upward because of higher Reynolds number.

Another uncertainty that comes due to unknown temperature while plotting the data is in the Reynolds number. Since the viscosity changes very rapidly with temperature, Reynolds number is actually affected inversely. Since the temperature of estimation of reported Reynolds number is unknown there could be a horizontal shift induced due to the Reynolds number. Hence if the viscosity would be estimated at higher temperature, the Reynolds number will be higher leading to a right side shift the experimental curve as well as theoretical curve.

Hence the experimental temperature seems to be an important factor in the estimation of HTC and comparison to experimental data. As discussed above a higher temperature leads to an upward shift of the theoretical estimation curve and right side shift of the experimental and theoretical HTC curve.

To illustrate the effect of viscosity variation with temperature on HTC, the data for viscosity obtained by Li et al [100] was modified to obtain viscosity at 45 C. This viscosity which was 67% of its room temperature value was then used to evaluate the Reynolds number and Prandtl number at 45 C and then
these were used in the Dittus Boelter correlation to predict the heat transfer coefficient. The new HTC obtained was plotted as a function of the new Reynolds number and compared to the results at 25°C. Figure 88 and Figure 89 show the comparison of results obtained. The dotted lines show the results obtained by assuming average experimental temperature of 45°C while the solid lines represent the results obtained by assuming 25°C as the temperature for calculation of nanofluid properties. As discussed above the experimental and theoretical curve shift towards right due to higher value of Reynolds number because of lower viscosity at higher temperature. And as shown in Table I, Nusselt number is higher which means a higher estimation of nanofluid HTC. Hence, it is clear from the above example that if the coolant temperature of 45°C would have been account for in viscosity, no anomalous enhancement should have been observed. Hence it could be possible to mistake the temperature effect on viscosity as anomalous heat transfer coefficient enhancement during turbulent flow. Similar trend was confirmed in all the 4 experimental studies analyzed for turbulent flows. Figures 90-93 confirm that trend.

![Effect of Temperature (2.78 vol% alumina)](image)

Figure 90 Pak et al
Effect of Temperature (3.16 vol% Titania)

Temperature Sensitivity of Data (0.2 vol% Titania)

Figure 91 Pak et al

Figure 92 Duangthongsuk et al
3.4.2. Laminar Flow

During this exercise, 8 laminar flow studies [93-100] were analyzed and most of the studies did display anomalous enhancement in the developing flow region. Another noticeable trend among most of the studies was that the enhancement was large at the entrance and decreased as the flow developed matching the theoretical prediction once the flow was fully developed. Interestingly, viscosity does not affect the prediction of HTC in the laminar flow regime as in case of turbulent flow. This is because of the fact that the laminar flow equations 16 and 17 account for Peclet number \((Pe = Re.Pr)\) hence viscosity is not a part of the prediction of HTC. The trend in the HTC in the developing regime is still remains unexplained and can be a good topic for future work.
4. Conclusion

An international nanofluid property benchmark exercise, or INPBE, was conducted by 34 organizations participating from around the world. The objective was to compare thermal conductivity data obtained by different experimental approaches for identical samples of various nanofluids. The main findings of the study were as follows:

- The thermal conductivity enhancement afforded by the tested nanofluids increased with increasing particle loading, particle aspect ratio and decreasing basefluid thermal conductivity.

- For all water-based samples tested, the data from most organizations deviated from the sample average by ±5% or less. For all PAO-based samples tested, the data from most organizations deviated from the sample average by ±10% or less.

- The classic effective medium theory for well-dispersed particles accurately reproduced the INPBE experimental data, thus suggesting that no anomalous enhancement of thermal conductivity was observed in the limited set of nanofluids tested in this exercise.

- Some systematic differences in thermal conductivity measurements were seen for different measurement techniques. However, as long as the same measurement technique at the same temperature conditions was used to measure the thermal conductivity of the basefluid, the thermal conductivity enhancement was consistent between measurement techniques.

Studies claiming an anomalous enhancement in HTC were analyzed and comparison was made with the theoretical estimation with proper calculation of nanofluid properties to be used in the prediction of nanofluid heat transfer coefficient. Uncertainties in the theoretical prediction/ experimental measurement of nanofluid properties were also accounted for while predicting HTC from theory. Also experimental uncertainties were estimated to the experimental HTC measurement. An enhancement beyond the
experimental/ theoretical uncertainties was confirmed as anomalous. The main trends observed in the analysis were

1) Experimentally measured HTC deviates from the theoretical prediction for nanofluids from about 10%-100%. Uncertainties in measurement of HTC were generally of the order of 5% and the theoretical estimation uncertainty was about 10%. Although enhancement during turbulent flow seems anomalous, it could be a case of mistaken temperature effect on viscosity as explained in Section 3.4. Anomalous enhancement could only be concluded for laminar flow.

2) A trend of higher deviation from the theoretical prediction at higher Reynolds number was seen. Although in case of turbulent flow this deviation could be solely due to temperature affect on viscosity, for laminar flow in developing regime this needs to be further investigated.

3) A trend of higher deviation from the theoretical prediction at higher concentration was observed suggesting an increasing HTC from factors other than just change in properties.

4) Analysis of temperature sensitivity of viscosity on the HTC prediction for turbulent flow shows that unknown fluid temperatures in experiments could lead to a significant shift in the theoretical prediction and experimental measurement curves as discussed in Section 3.4. Hence use of viscosity corresponding to the correct fluid temperature is very important during turbulent flow and could be a possible reason for the observed anomalous enhancement in the turbulent flow studies. However the temperatures of experiments need to be confirmed from the researchers.

5) Interestingly estimation of viscosity doesn’t affect the prediction of laminar flow HTC because theoretical correlations always account for Peclet number ($Pe=Re.Pr$) which eliminates viscosity.

6) An anomalous enhancement was systematically observed during laminar flow in the entrance regime. This enhancement in the laminar flow during developing flow regime couldn’t be
explained. Possible sources of such an enhancement still need to be explored and can be extended as future work.
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Appendix A - Statistical treatment of INPBE data

For each fluid sample, the thermal conductivity raw data \((x_{i1}, x_{i2}, ..., x_{iN})\) from the \(i\)-th organization were processed to estimate the organizational mean \((\bar{x}_i)\) and standard deviation \((s_i)\), respectively, as:

\[
\bar{x}_i = \frac{1}{n_i} \sum_{j=1}^{n_i} x_{ij} \quad \text{and} \quad s_i = \sqrt{\frac{1}{n_i-1} \sum_{j=1}^{n_i} (x_{ij} - \bar{x}_i)^2}
\]  

(A.1)

The values of \(\bar{x}_i\) and \(s_i\) for each organization are shown in Figures 6 through 18 as data points and error bars, respectively. The normality of the \(x_y\) datasets was checked using the Shapiro-Francia W' test\(^{56}\) and was found to be satisfactory. Peirce's criterion\(^{57}\) was used to identify outliers which were not included in the sample average and variance calculations described below, but are shown in Figures 6 – 18 as filled data points.

The analysis of data among different organizations was carried out using the Random Effects Model\(^{56}\). In the Random Effects Model, an assumption is made that the conclusions from the analysis can be applied to a wider class of measurements of which the \(n_i\) populations (or organizations, in this case) are a representative subset. The model assumes that:

\[
x_{ij} = \mu + \alpha_i + e_{ij}
\]

(A.2)

where \(\mu\) is the estimator of the sample mean, \(\alpha_i\) is the systematic error for each organization (which are treated as random errors among organizations), and \(e_{ij}\) is the random or unexplained error for each measurement. It is helpful to note that

\[
\alpha_i = \bar{x}_i - \mu
\]

\[
e_{ij} = x_{ij} - \bar{x}_i
\]

(A.3)

It is assumed that \(\alpha_i\) and \(e_{ij}\) are normally distributed with zero means and standard deviations of \(\sigma_\alpha\) and \(\sigma_e\), respectively. The normality of the \(e_{ij}\) datasets was checked using the Shapiro-Francia W' test and was
found to be satisfactory. This analysis assumes that standard deviations within the organizations are equal
($\sigma_i = \sigma_e$). This was checked by performing pair-wise F-tests on $\sigma_i$.

The standard Random Effects Model uses a weighted average as the sample average (taking into account
the number of data points reported by each organization).

$$
\mu = \bar{x} = \frac{1}{N} \sum_{i=1}^{l} n_i \bar{x}_i
$$

(A.4)

We believe that this definition overemphasizes the contributions from organizations that reported many
data points. For the purposes of this study, a more appropriate estimator of the sample mean is an
unweighted average of organization averages given in the following equation:

$$
\bar{x} = \frac{1}{l} \sum_{i=1}^{l} \bar{x}_i
$$

(A.5)

This way, each organization contributes equally to the ensemble average. This estimator has been
analyzed in the literature and its variance is given by

$$
\sigma^2 = Var(\bar{x}) = \frac{1}{l^2} \sum_{i=1}^{l} \left( \frac{\sigma^2}{n_i} + \sigma_e^2 \right)
$$

(A.6)

where

$$
\sigma_e^2 = \frac{1}{(N - l)} \sum_{i=1}^{l} (n_i - 1)s_i^2
$$

(A.7)

$$
N = \sum_{i=1}^{l} n_i
$$

(A.8)

$$
\sigma_o^2 = \frac{(MSA - \sigma_e^2)}{n_o}
$$

(A.9)
\[ MSA = \frac{1}{(I-1)} \left( \sum_{i=1}^{I} n_i \bar{x}_i^2 - N \bar{x}^2 \right) \]  \hspace{1cm} (A.10)

\[ n_o = \frac{1}{I-1} \left( \frac{\sum_{i=1}^{I} n_i^2}{N - \frac{\sum_{i=1}^{I} n_i}{N}} \right) \]  \hspace{1cm} (A.11)

The standard error (\(\sqrt{\sigma}\)) of the unweighted average is shown in Figures 6 – 18 as dotted lines plotted above and below the sample average. The literature shows that the estimator (A.5) is preferred over the estimator (A.4) if \(\sigma^2_0 > \sigma^2_e\) \(^{56,58}\). The statistical analysis shows that this condition is satisfied for the INPBE data.

Thermal conductivity ratios were determined from the ratio of the nanofluid thermal conductivity to the basefluid thermal conductivity and are given as \(y_i\). If an organizational mean for a given fluid sample was identified as an outlier in the thermal conductivity analysis, it was not excluded here in determining enhancements. A second round of applying the Peirce criterion excluded those enhancements that were outliers.

The standard deviation (error bars) of the thermal conductivity enhancements (data points) for individual organizations shown in Figures 19 through 26 were calculated by propagating the standard deviation of the numerator and denominator \(^{59}\). That is, if \(y = \frac{x_{nf}}{x_{bf}}\) then:

\[ \frac{S_{enh}}{y} = \sqrt{\left( \frac{S_{nf}}{x_{nf}} \right)^2 + \left( \frac{S_{bf}}{x_{bf}} \right)^2} \]  \hspace{1cm} (A.12)

The procedure for calculating the thermal conductivity enhancement sample average and its variance was based on Eqs. (A.5) through (A.11), where the thermal conductivity for each organization, \(\bar{x}_i\), is replaced
by the thermal conductivity enhancement for each organization, $\overline{y}_i$, and $n_i$ is the harmonic average of the total number of measurements used to calculate the enhancement.

To compare the different measurement techniques, the Fixed Effects Model was used. For each technique, the technique average and the variance were determined using Eqs. (A.5) through (A.11) above. For an unbalanced data set (one in which there are a different number of data points for each measurement technique to be compared), the approximate Tukey-Kramer intervals were used, which depend on the probability statement,

$$P\left\{\mu_i - \mu_{i'} \in \overline{y}_i - \overline{y}_{i'} \pm q_{\alpha,k,v} s_i \sqrt{\frac{1}{n_i} + \frac{1}{n_{i'}}}\right\}^{1/2} = 1 - \alpha , \quad \text{(A.13)}$$

where $q_{\alpha,k,v}$ is the upper $\alpha$ point of the "studentized" range distribution for $k$ (the number of measurement techniques compared) and $v$, the degrees of freedom ($N - k$). If the interval given in Eq. (A.12) does not contain zero for any combination of two measurement techniques, then the difference in technique mean is statistically significant.
APPENDIX B- Error Analysis for Theoretical Heat Transfer Coefficient

The error analysis for the HTC was done using the standard propagation of error. Prediction of HTC using theory requires the knowledge of following thermal hydraulic variables: thermal conductivity ($k$), viscosity ($\mu$), density ($\rho$), specific heat ($C_p$), flowrate ($v$) and temperature ($T$). Hence

\[ h = f(k, \rho, \mu, C_p, v, T) \] \hspace{1cm} B.1

Neglecting the temperature effect, the variance in the heat transfer coefficient predicted using the values of these thermal hydraulic variables was calculated using the standard error propagation given by equation B.2

\[ \sigma_h^2 = \left( \frac{\partial h}{\partial k} \sigma_k \right)^2 + \left( \frac{\partial h}{\partial \rho} \sigma_\rho \right)^2 + \left( \frac{\partial h}{\partial \mu} \sigma_\mu \right)^2 + \left( \frac{\partial h}{\partial C_p} \sigma_{C_p} \right)^2 + \left( \frac{\partial h}{\partial v} \sigma_v \right)^2 \] \hspace{1cm} B.2

where $\sigma_X$ represents the standard deviation in quantity $X$. The partial derivate w.r.t. variable $X$ is calculated by differentiating the correlation used for the prediction of HTC w.r.t. variable $X$. $\sigma_X$ was obtained from the experimentally reported values or was estimated assuming standard uncertainties.

The standard deviation in HTC data is obtained from equation B.2 by taking the square root of the variance. This is value which is plotted on the error bars of theoretical estimated HTC. The error bars in the figures are $2\sigma_h$ long spanning $\sigma_h$ in both directions. Hence we are 66.66% sure that our estimation is within these error bounds. Hence, a significant deviation beyond this bound even after accounting for experimental measurement uncertainty can be safely termed as anomalous.

In case a thermo-physical property of nanofluid was estimated using theoretical models, the uncertainty in the estimation of that property was estimated in the same way using standard propagation of errors through the independent variables (which were properties of nanoparticles and basefluid).