Experiments on Laminar Convective Heat Transfer

with $\gamma$-$\text{Al}_2\text{O}_3$ Nanofluids

by

Wei-Yun Lai

A Dissertation Presented in Partial Fulfillment
of the Requirements for the Degree
Doctor of Philosophy

Approved October 2010 by the
Graduate Supervisory Committee:

Patrick E Phelan, Chair
Kangping Chen
Mark Hayes
Ravi S Prasher
Karl Sieradzki

ARIZONA STATE UNIVERSITY

December 2010
ABSTRACT

As miniature and high-heat-dissipation equipment became major manufacture and operation trends, heat-rejecting and heat-transport solutions faced increasing challenges. In the 1970s, researchers showed that particle suspensions can enhance the heat transfer efficiency of their base fluids. However, their work was hindered by the sedimentation and erosion issues caused by the relatively large particle sizes in their suspensions. More recently, nanofluids—suspensions of nanoparticles in liquids—were proposed to be applied as heat transfer fluids, because of the enhanced thermal conductivity that has generally been observed. However, in practical applications, a heat conduction mechanism may not be sufficient for cooling high-heat-dissipation devices such as microelectronics or powerful optical equipment. Thus, the thermal performance under convective, i.e., flowing heat transfer conditions becomes of primary interest. In addition, with the presence of nanoparticles, the viscosity of a nanofluid is greater than its base fluid and deviates from Einstein’s classical prediction. Through the use of a test rig designed and assembled as part of this dissertation, the viscosity and heat transfer coefficient of nanofluids can be simultaneously determined by pressure drop and temperature difference measurements under laminar flow conditions. An extensive characterization of the nanofluid samples, including pH, electrical conductivity, particle sizing and zeta potential, is also documented. Results indicate that with constant wall heat flux, the
relative viscosities of nanofluid decrease with increasing volume flow rate. The results also show, based on Brenner's model, that the nanofluid viscosity can be explained in part by the aspect ratio of the aggregates. The measured heat transfer coefficient values for nanofluids are generally higher than those for base fluids. In the developing region, this can be at least partially explained by Prandtl number effects. The Nusselt number \((Nu)\) results for nanofluid show that \(Nu\) increases with increasing nanofluid volume fraction and volume flow rate. However, only DI-H\(_2\)O (deionized water) and 5/95 PG/H\(_2\)O (PG = propylene glycol) based nanofluids with 1 vol% nanoparticle loading have \(Nu\) greater than the theoretical prediction, 4.364. It is suggested that the nanofluid has potential to be applied within the thermally developing region when utilizing the nanofluid as a heat transfer liquid in a circular tube. The suggested Reynold’s number is greater than 100.
To my family.
ACKNOWLEDGEMENTS

First and foremost, I would like to thank my family for providing an abundance of love, encouragement and support along my lengthy and sometimes wandering path throughout the years of my pursuit of the doctoral degree.

I would also like to dedicate special thanks to my advisor, Prof. P.E. Phelan, who has provided considerable wealth of resources, opportunities and guidance during my study of nanofluids. His support has been an essential contributing factor towards the completion of this manuscript. I would also like to thank the rest of my committee, Prof. Kangping Chen, Prof. Mark Hayes, Dr. Ravi Prasher, and Prof. Karl Sieradzki, who have been very generous with their time and insights. Other faculty members of the graduate school whose guidance I highly appreciate includes Prof. Ronald Calhoun, Prof. Taewoo Lee and Prof. Jonathan Posner.

I am grateful for the financial support from National Science Foundation, through a GOALI award (Award No. CTS-0353543), ASU University Graduate Fellowship, Graduate Research and Teaching Assistantships, and the direct support provided by the Intel Corporation.

My stay at ASU has been made so unforgettable and enjoyable by all the people I have worked with through these years. I am indebted to Dr. Lynn Cozort and Moya Gilchrist for their generous assistance. I would like to thank Dr. Bruce Steele, Prajesh Bhattacharya, Yeshpal Gupta, Brent
Odom, Rob Taylor, Himanshu Tyagi, Sabarish Vinod, Mark Witt and Moxuan Zhu for their friendship and technical support.
# TABLE OF CONTENTS

| LIST OF TABLES | ................................................................. | x |
| LIST OF FIGURES | ................................................................. | xi |
| NOMENCLATURE | ................................................................. | xix |

## CHAPTER

1. INTRODUCTION ................................................................. 1
   1.1. Investigations for thermal conductivity enhancement of nanofluid ................................................................. 3
   1.2. Investigations of nanofluid viscosity ................................................................. 5
   1.3. Investigations of nanofluids convective heat transfer ................................................................. 6
   1.4. Research objectives ................................................................. 8

2. EXPERIMENTAL METHODOLOGY AND APPARATUS
   VALIDATION OF NANOFLUID VISCOSITY AND CONVECTIVE HEAT TRANSFER COEFFICIENT ................................................................. 11
   2.1. Experimental system for measuring nanofluid viscosity and convective heat transfer coefficient ................................................................. 11
   2.2. Data analysis for nanofluid viscosity ................................................................. 15
   2.3. Data analysis for nanofluid convective heat transfer coefficient ................................................................. 16
   2.4. Calibration and uncertainty analysis ................................................................. 17
   2.5. Apparatus validation ................................................................. 18
   2.6. Summary ................................................................. 21
# CHAPTER 3. NANOFLUID PREPARATION, CHARACTERIZATION AND STABILITY

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.1. Nanofluid preparation</td>
<td>23</td>
</tr>
<tr>
<td>3.2. Size and shape of dry nanoparticles</td>
<td>25</td>
</tr>
<tr>
<td>3.3. Nanofluid characterization</td>
<td>29</td>
</tr>
<tr>
<td>3.3.1. Nanoparticle size in suspension</td>
<td>29</td>
</tr>
<tr>
<td>3.3.2. Nanofluid pH, electrical conductivity and zeta potential</td>
<td>30</td>
</tr>
<tr>
<td>3.3.3. Results of nanofluid characterization</td>
<td>33</td>
</tr>
<tr>
<td>3.4. Nanofluid stability</td>
<td>45</td>
</tr>
<tr>
<td>3.4.1. Calculation of nanofluid stability ratio</td>
<td>45</td>
</tr>
<tr>
<td>3.4.2. Results of nanofluid stability ratio</td>
<td>47</td>
</tr>
<tr>
<td>3.5. Summary</td>
<td>49</td>
</tr>
</tbody>
</table>

# CHAPTER 4. PRESSURE-DROP VISCOSITY MEASUREMENT $\gamma$-Al$_2$O$_3$ NANOPARTICLES IN WATER AND PROPYLENE GLYCOL-WATER MIXTURE

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.1. Literature review</td>
<td>50</td>
</tr>
<tr>
<td>4.2. Viscosity of base fluids</td>
<td>57</td>
</tr>
<tr>
<td>4.3. Results and discussion of nanofluid viscosity data</td>
<td>59</td>
</tr>
<tr>
<td>4.4. Summary</td>
<td>68</td>
</tr>
<tr>
<td>CHAPTER</td>
<td>Page</td>
</tr>
<tr>
<td>---------</td>
<td>------</td>
</tr>
<tr>
<td>5. CONVECTIVE HEAT TRANSFER COEFFICIENT OF ALUMINA NANOFLUIDS</td>
<td>69</td>
</tr>
<tr>
<td>5.1. Convective heat transfer results of DI-H₂O based nanofluid</td>
<td>76</td>
</tr>
<tr>
<td>5.2. Convective heat transfer results of 5/95 PG/H₂O based nanofluid</td>
<td>79</td>
</tr>
<tr>
<td>5.3. Convective heat transfer results of 15/85 PG/H₂O based nanofluid</td>
<td>83</td>
</tr>
<tr>
<td>5.4. Discussion</td>
<td>86</td>
</tr>
<tr>
<td>5.5. Summary</td>
<td>88</td>
</tr>
<tr>
<td>6. NANOFLUID CONVECTIVE HEAT TRANSFER PERFORMANCE IN NONDIMENSIONAL FORM IN FULLY DEVELOPED REGION</td>
<td>89</td>
</tr>
<tr>
<td>6.1. Calculation of nanofluid thermal conductivity</td>
<td>89</td>
</tr>
<tr>
<td>6.1.1. Thermal conductivity model</td>
<td>89</td>
</tr>
<tr>
<td>6.1.2. Results of nanofluid thermal conductivity</td>
<td>92</td>
</tr>
<tr>
<td>6.2. Nu results in the thermally developing region</td>
<td>96</td>
</tr>
<tr>
<td>6.3. Nu results in the fully developed region</td>
<td>102</td>
</tr>
<tr>
<td>6.4. Discussion</td>
<td>105</td>
</tr>
<tr>
<td>6.5. Summary</td>
<td>114</td>
</tr>
<tr>
<td>7. CONCLUSIONS AND FUTURE WORK</td>
<td>115</td>
</tr>
<tr>
<td>7.1. Conclusions</td>
<td>115</td>
</tr>
<tr>
<td>7.2. Future work</td>
<td>117</td>
</tr>
</tbody>
</table>
A MODIFIED HAMAKER CONSTANT OF BINARY LIQUID MIXTURE

................................................................. 129
LIST OF TABLES

<table>
<thead>
<tr>
<th>Table</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.1. Properties of base fluids at 25°C</td>
<td>24</td>
</tr>
<tr>
<td>3.2. pH and electrical conductivity of alumina nanoparticles in different base fluids</td>
<td>35</td>
</tr>
<tr>
<td>3.3. Hamaker constant $^1$ ($A_{131}$) and stability ratio of this study</td>
<td>48</td>
</tr>
<tr>
<td>4.1. Summary of experimental studies on nanofluid viscosity</td>
<td>52</td>
</tr>
<tr>
<td>4.2. Summary of measured base fluid viscosities (all viscosity units are in mPa·s)</td>
<td>58</td>
</tr>
<tr>
<td>5.1. Summary of experimental studies on convective heat transfer performance of nanofluid under laminar flow region</td>
<td>71</td>
</tr>
<tr>
<td>6.1. Aggregation time constant of each nanofluid using eq. (6.9)</td>
<td>93</td>
</tr>
</tbody>
</table>
LIST OF FIGURES

<table>
<thead>
<tr>
<th>Figure</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.1. Schematic diagram of experimental apparatus for convective heat transfer and viscosity measurement</td>
<td>12</td>
</tr>
<tr>
<td>2.2. Sketch of heater wire and thermocouple locations (not drawn to scale)</td>
<td>12</td>
</tr>
<tr>
<td>2.3. Cross-sectional view of test tube (not drawn to scale)</td>
<td>12</td>
</tr>
<tr>
<td>2.4. Pressure drop across the test section using pure DI-H₂O (no nanoparticles) as the working fluid</td>
<td>19</td>
</tr>
<tr>
<td>2.5. Comparison between measured and calculated [61] heat transfer coefficients for developing flow, for pure DI-H₂O (no nanoparticles)</td>
<td>19</td>
</tr>
<tr>
<td>3.1. Transmission electron microscope (TEM) image for 20-nm dry Al₂O₃ nanopowder. The clusters show size ranges from hundred nm to a few μm. The bright gray oval spots are sample-holding substrate. The dark black spots are due to strong agglomeration of dry nanoparticles</td>
<td>26</td>
</tr>
<tr>
<td>3.2. (a) TEM image for dry Al₂O₃ nanopowder. The image shows clusters of spherical, nearly spherical and some arbitrary shaped nanoparticles</td>
<td>28</td>
</tr>
<tr>
<td>3.2. (b) TEM image for 1 vol% Al₂O₃-water after water evaporated</td>
<td>28</td>
</tr>
</tbody>
</table>
3.3. Example of a good power spectrum during zeta potential measurements for determining nanofluids concentrations for DLS measurement. The red and black curves are the reference and the sample relative frequencies .............................................. 32

3.4. Zeta potential vs time after sonication for diluted Al₂O₃ nanofluids at different original (undiluted) volume fractions ...... 36

3.5. An example of a particle size distribution measured with the Nicomp 380/ZLS system. Screen shot is the particle sizing result of diluted 20-nm γ-Al₂O₃ nanoparticles in DI- H₂O taken 70 minutes after sonication. The distribution is generated by the Nicomp volume-weighted model.............................................38

3.6. (a) Diluted aggregate mean diameters and % of volume vs. original nanoparticle volume fraction 60 mins after sonication (DI-H₂O)......................................................................................... 39

3.6. (b) Diluted aggregate mean diameters and % of volume vs. original nanoparticle volume fraction 70 mins after sonication (DI-H₂O).........................................................................................40

3.6. (c) Diluted aggregate mean diameters and % of volume vs. original nanoparticle volume fraction 80 mins after sonication (DI-H₂O).........................................................................................40
<table>
<thead>
<tr>
<th>Figure</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.7. (a) Diluted aggregate mean diameters and %volume vs. original nanoparticle volume fraction 60 mins after sonication (5/95 PG/H₂O)</td>
<td>42</td>
</tr>
<tr>
<td>3.7. (b) Diluted aggregate mean diameters and %volume vs. original nanoparticle volume fraction 70 mins after sonication (5/95 PG/H₂O)</td>
<td>43</td>
</tr>
<tr>
<td>3.7. (c) Diluted aggregate mean diameters and %volume vs. original nanoparticle volume fraction 80 mins after sonication (5/95 PG/H₂O)</td>
<td>43</td>
</tr>
<tr>
<td>3.8. (a) Diluted aggregate mean diameters and %volume vs. original nanoparticle volume fraction 60 mins after sonication (15/85 PG/H₂O)</td>
<td>44</td>
</tr>
<tr>
<td>3.8. (b) Diluted aggregate mean diameters and %volume vs. original nanoparticle volume fraction 70 mins after sonication (15/85 PG/H₂O)</td>
<td>44</td>
</tr>
<tr>
<td>3.8. (c) Diluted aggregate mean diameters and %volume vs. original nanoparticle volume fraction 80 mins after sonication (15/85 PG/H₂O)</td>
<td>45</td>
</tr>
<tr>
<td>4.1. Experimental values of base fluid viscosities</td>
<td>58</td>
</tr>
<tr>
<td>4.2. Experimental results of relative viscosity of 20-nm Al₂O₃-H₂O nanofluids with different volume fraction. The ‘H’ curves represent measurements under heated wall conditions</td>
<td>60</td>
</tr>
</tbody>
</table>
4.3. Experimental results of relative viscosity of 20-nm Al$_2$O$_3$ nanoparticles in 5/95 PG/H$_2$O with different volume fraction.....60

4.4. Experimental results of relative viscosity of 20-nm Al$_2$O$_3$ nanoparticles in 15/85 PG/H$_2$O with different volume fraction... 61

4.5. Experimental values for 20-nm Al$_2$O$_3$ nanoparticles in different base fluids for a volume flow rate of 5 mL/min. The ‘H’ curves represent measurements under heated wall conditions ...............62

4.6. Comparison of 20-nm Al$_2$O$_3$-H$_2$O experimental values with Einstein’s model [31] and the Prasher et al. experimental correlation [27]. Relative viscosities of long thin prolate spheroids predicted by Brenner [86] are also included. The ‘H’ data represent measurements under heated wall conditions, and ‘$a_p$’ is the particle axis ratio................................................................. 65

5.1. (a) Local heat transfer coefficients of DI-H$_2$O based nanofluids in the thermal developing region at 1 mL/min volume flow rate................................................................. 77

5.1. (b) Local heat transfer coefficients of DI-H$_2$O based nanofluids in the thermal developing region at 5 mL/min volume flow rate................................................................. 77

5.1. (c) Local heat transfer coefficients of DI-H$_2$O based nanofluids in the thermal developing region at 9 mL/min volume flow rate................................................................. 78
<table>
<thead>
<tr>
<th>Figure</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.2. Heat transfer coefficient of water-based 20-nm Al$_2$O$_3$ nanofluids in the fully developed region</td>
<td>78</td>
</tr>
<tr>
<td>5.3. (a) Local heat transfer coefficients of 5/95 PG/H$_2$O based nanofluids in the thermal developing region at 1 mL/min volume flow rate</td>
<td>80</td>
</tr>
<tr>
<td>5.3. (b) Local heat transfer coefficients of 5/95 PG/H$_2$O based nanofluids in the thermal developing region at 5 mL/min volume flow rate</td>
<td>81</td>
</tr>
<tr>
<td>5.3. (c) Local heat transfer coefficients of 5/95 PG/H$_2$O based nanofluids in the thermal developing region at 9 mL/min volume flow rate</td>
<td>81</td>
</tr>
<tr>
<td>5.4. Heat transfer coefficient of 5/95 PG/H$_2$O based 20-nm Al$_2$O$_3$ nanofluids in the fully developed region</td>
<td>82</td>
</tr>
<tr>
<td>5.5. (a) Local heat transfer coefficients of 15/85 PG/H$_2$O based nanofluids in the thermal developing region at 1 mL/min volume flow rate</td>
<td>84</td>
</tr>
<tr>
<td>5.5. (b) Local heat transfer coefficients of 15/85 PG/H$_2$O based nanofluids in the thermal developing region at 5 mL/min volume flow rate</td>
<td>84</td>
</tr>
<tr>
<td>5.5. (c) Local heat transfer coefficients of 15/85 PG/H$_2$O based nanofluids in the thermal developing region at 9 mL/min volume flow rate</td>
<td>85</td>
</tr>
</tbody>
</table>
5.6. Heat transfer coefficient of 15/85 PG/H₂O based 20-nm Al₂O₃ nanofluids in the fully developed region ..................................................85

6.1. Schematic diagram of well dispersed clusters. \( R_a \) is the gyration radius of aggregates and the red curve indicates high thermal conductivity-percolation path ........................................92

6.2. Thermal conductivity ratio of DI-H₂O based nanofluids w.r.t. time. The band indicates the period when convection experiments were conducted ..............................................................................94

6.3. Thermal conductivity ratio of 5/95 PG/H₂O based nanofluids. Band indicates the period when convection experiments were conducted ..........................................................................................95

6.4. Thermal conductivity ratio of 15/85 PG/H₂O based nanofluids. Band indicates the period when convection experiments were conducted ..........................................................................................95

6.5 (a). \( Nu \) vs. \( x/L \) of DI-H₂O based nanofluids with Al₂O₃ nanoparticles, for thermally developing flow with \( Re \sim 25 \) ............ 97

6.5 (b). \( Nu \) vs. \( x/L \) of DI-H₂O based nanofluids with Al₂O₃ nanoparticles, for thermally developing flow with \( Re \sim 120 \) ............ 97

6.5 (c). \( Nu \) vs. \( x/L \) of DI-H₂O based nanofluids with Al₂O₃ nanoparticles, for thermally developing flow with \( Re \sim 210 \) ............ 98

6.6 (a). \( Nu \) vs. \( x/L \) of 5/95 PG/H₂O based nanofluids with Al₂O₃ nanoparticles, for thermally developing flow with \( Re \sim 20 \) ............ 99
6.6 (b). Nu vs \( x/L \) of 5/95 PG/H\(_2\)O based nanofluids with Al\(_2\)O\(_3\) nanoparticles, for thermally developing flow with \( Re \approx 110 \)...........99

6.6 (c). Nu vs \( x/L \) of 5/95 PG/H\(_2\)O based nanofluids with Al\(_2\)O\(_3\) nanoparticles, for thermally developing flow with \( Re \approx 200 \)........100

6.7 (a). Nu vs \( x/L \) of 15/85 PG/H\(_2\)O based nanofluids with Al\(_2\)O\(_3\) nanoparticles, for thermally developing flow with \( Re \approx 10 \) ..........100

6.7 (b). Nu vs \( x/L \) of 15/85 PG/H\(_2\)O based nanofluids with Al\(_2\)O\(_3\) nanoparticles, for thermally developing flow with \( Re \approx 55 \) ..........101

6.7 (c). Nu vs \( x/L \) of 15/85 PG/H\(_2\)O based nanofluids with Al\(_2\)O\(_3\) nanoparticles, for thermally developing flow with \( Re \approx 100 \) ..........101

6.8. Nu vs. \( Re \) of DI-H\(_2\)O based nanofluids with Al\(_2\)O\(_3\) nanoparticles .......................................................... 103

6.9. Nu vs. \( Re \) of 5/95 PG/H\(_2\)O based nanofluids with Al\(_2\)O\(_3\) nanoparticles .......................................................... 103

6.10. Nu vs. \( Re \) of 15/85 PG/H\(_2\)O based nanofluids with Al\(_2\)O\(_3\) nanoparticles .......................................................... 104

6.11. Aggregation size vs. time of 0.5 vol\% and 0.75 vol\% Al\(_2\)O\(_3\) in PG/H\(_2\)O mixtures .......................................................... 107
6.12. Comparison of thermal conductivity ratio by different aggregation time constant. Black lines are results by pure theoretical model of [62] and green lines indicate the results from combined experimental/ theoretical predictions. Nanofluids are 0.5 vol% and 0.75 vol% Al₂O₃ in 5/95 PG/H₂O base fluids.

6.13. Comparison of thermal conductivity ratio by different aggregation time constant. Black lines are results by pure theoretical model of [62] and green lines indicate the results from combined experimental/ theoretical predictions. Nanofluids are 0.5 vol% and 0.75 vol% Al₂O₃ in 15/85 PG/H₂O base fluids.

6.14. Comparisons of \( Nu \) vs. \( Re \) using modified and non-modified \( k \) of PG/H₂O based nanofluids. The blue line indicates the constant \( Nu \) result from the traditional analytical solution. The closed symbols are the \( Nu \) results when using \( k \) results by combined experimental/theoretical approach and open symbols are \( Nu \) when using \( k \) results by predictions of [62].
NOMENCLATURE

$A$  Area [m$^2$]
$A_{131}$  Hamaker constant [J]
$a$  Length of polar radius [m]
$a_p$  Particle axial ratio [-]
$B$  Particle hydrodynamic interaction correction factor [-]
$b$  Length of equatorial radius
$C$  Enhancement coefficient [-]
$c$  Heat capacity per unit volume [J m$^{-3}$ K$^{-1}$]
$D$  Diameter [m]
$d_f$  Fractal dimension [-]
$F$  Faraday’s constant $9.649 \times 10^4$ [C mol$^{-1}$]
$h$  Convective heat transfer coefficient [W m$^{-2}$ K$^{-1}$]
$\hat{h}$  Plank constant $6.626 \times 10^{-34}$ [J s]
$I$  Current [A]
$I_s$  Ionic strength [mol L$^{-1}$]
$k$  Thermal conductivity [W m$^{-1}$ K$^{-1}$]
$k_B$  Boltzmann Constant [J K$^{-1}$]
$L$  Length [m]
$n$  Refractive index [-]
$P$  Pressure [Pa]
$Q$  Heat rate [W]
$q''$  Heat flux [W m$^{-2}$]
$R_a$  Gyration radius [m]
$R_c$  Universal gas constant $8.314$ [J mol$^{-1}$ K$^{-1}$]
$r$  Radius [m]
$s$  Particle separation distance [m]
$T$  Temperature [K]
$t$  Time [s]
$t_p$  Aggregation time constant [s]
$U$  Velocity [m s$^{-1}$]
$V$  Voltage [V]
$\dot{V}$  Volume flow rate [mL min$^{-1}$]
$V_A$  Attraction potential energy [J]
$V_R$  Repulsion potential energy [J]
$\nu_e$  Plasma frequency [s$^{-1}$]
$W$  Stability ratio [-]
$x$  axial direction [-]

Greek Symbols
$\delta$  Hydraulic boundary thickness [m]
$\delta_t$  Thermal boundary layer thickness [m]
$\varepsilon_0$  Dielectric permittivity of a vacuum $8.854 \times 10^{-12}$ [F m$^{-1}$]
$\varepsilon_r$  Relative dielectric permittivity [-]
$\eta$ Electrophoretic mobility [cm$^2$Vs]
$k$ Debye-Hückel parameter [m$^{-1}$]
$\mu$ Viscosity [Pa s]
$\sigma$ Electrical conductivity [$\mu$S cm$^{-1}$]
$\tau$ Shear stress [s$^{-1}$]
$\phi$ Particle volume fraction [-]
$\rho$ Density [kg m$^{-3}$]
$\zeta$ Zeta potential [mV]

Subscripts

$a$ Aggregate
$b$ Base fluid
$eff$ Effective
$f$ Fluid
$i$ Inner
$m$ Mean
$nf$ Nanofluid
$o$ Outer
$p$ Particle
$w$ Wall
Chapter 1: Introduction

The transport of heat has been a recognized challenge in human history. For instance, indoor temperature control for the human body, food preservation in any weather conditions, rejecting heat from computer chips or optical devices, regenerators of gas-turbine engines, cooling systems for power plants, etc., are examples of heat-exchange processes, which either input heat into a system or remove heat produced in a system. These heat transfer applications directly or indirectly affect people’s daily life and continue to require additional research in order to improve their efficiency. Responding to these needs, researchers have introduced various enhancement methods since the 1950s. In 1959, Nobel Prize winner Richard Feynman introduced the concept of micromachines at the annual meeting of the American Physical Society in his famous speech “There’s plenty of room at the bottom” [1]. Ever since then, the understanding of the physics behind the microscale and nanoscale domains has become a major research trend. Along with an advancement in manufacturing techniques, the products that have small size, high-heat-flux and non-uniform heat flux have occupied a significant portion in many industries. This trend is expected to continue unabated for the coming years and therefore heat-rejecting solutions are facing strict demands nowadays.

From an engineering point of view, forced convection utilizing liquid coolants in laminar or turbulent flow regimes are always a key heat
transport solution for the examples stated above [2]. A higher heat transfer coefficient means better convective performance. Several enhancement techniques were listed in [2], including heat transfer surface roughness modification, extended surfaces using fins, surface vibration, injection, and so on. However, these techniques usually lead to dramatically higher pressure loss and hence increase pumping power requirement. Also, with low thermal conductivity and/or high viscosity of common heat transfer liquids like water, ammonia, ethylene glycol, and mineral oil, the convective thermal performance was often inefficient and created barriers in designing small heat rejecting devices. Therefore, an innovative coolant with improved heat transfer properties is desired.

Since solid particles usually exhibit greater thermal conductivity than do liquids, one approach for improving the thermal conductivity of liquids is by using suspensions, which contain dispersed particles into base fluids. Pioneering researches of stationary, dilute, dispersions of solid spheres were conducted and published by James Clark Maxwell [3]. This work was extended further but much later to examine polystyrene suspensions by Ahuja [4, 5] in 1975. Ahuja conducted a series of experiments of thermal conductivity and heat transfer coefficients of 40-100 μm-sized polystyrene-water based solutions in a 1-mm-i.d. tube. It was shown that, without significant pressure loss, the effective thermal conductivity of the colloidal solution was improved by as much as a factor of 2. Furthermore, as the Reynolds number and particle concentration
increased, so did the enhancement in the effective thermal conductivity of the suspension. Ahuja proposed the enhancement in heat transfer coefficient is due to particle rotation by the shear field of tube flow.

However, the size of the particles was correspondingly large (in micro-scale) because a lack of available technology in those years. Owing to the size, several penalties when using micro-particle or larger particle suspensions are (1) the particles are not stable enough and have a tendency to settle out of suspension; and (2) larger particles can easily cause erosion to flow loop components. And hence, even though such large suspensions promise enhanced cooling ability, liquids consisting of such coarse-grained particles have not yet been widely applied as alternative coolants.

1.1. Investigations for thermal conductivity enhancement of nanofluids

With rapid development in manufacturing techniques and nanotechnology, the production of nanoparticles (particle size less than 100 nm) has been made possible. Nanofluids are suspended nanoparticles or fibers in heat transfer liquids, which were conceived by Choi [6] in 1995. The first impression of nanofluid is the observation of greater-than-expected effective thermal conductivity with small nanoparticle volume fraction [6, 7]. Since then, researchers have shown considerable interest in using nanoparticles as additives to alter heat transfer fluids and their thermal performance [8-16].
By virtue of the large number of ongoing investigations, a number of review articles have been published on nanofluids [17-26], with most of the emphasis placed on observed and predicted enhancements in the effective thermal conductivity. Explanations offered for these abnormal behaviors in the literature included surface roughness effect, chaotic movement (Brownian motion) and particle-particle interactions of suspended nanoparticles, high specific area which increases heat transfer surface between fluids and particles, and particle coagulation which creates high-conductivity heat transfer paths. In an effort to reduce the experimental artifacts caused by the traditional “hot-wire” approach to measure nanofluid thermal conductivity, some groups adopted optical measurement methods, and did not observe great enhancements in the effective thermal conductivity [13-15]. Putnam et al. [13] observed that the effective static thermal conductivities of gold (Au) nanofluids were independent of particle loading. Venerus et al. [14] studied the thermal conductivity of Au and alumina oxide (Al$_2$O$_3$) nanofluids via the forced Rayleigh scattering technique. The thermal conductivity data from their measurements were independent of temperature and the enhancements were consistent with the predictions from the classical effective medium theory. Furthermore, measurements on monodisperse polymer suspensions by Rusconi et al. [15] did not show an anomalous increase in thermal conductivity, but rather followed the classical models for the effective properties of composite media.
1.2. Investigations of nanofluid viscosity

Since the nanofluid convective heat transfer performance depends on the compromise between the increment of thermal conductivity and the increment of viscosity [27], it is critical to study nanofluid viscosity when a system employs fluid flow. The viscosity of a fluid is expected to increase as particles are added, no matter if the particles are rotating or non-rotating in the flow field [28]. In the test flow regime of this study--laminar flow--the pressure drop along a tube is directly proportional to viscosity. This increment in viscosity will lead to an undesired increase in pumping power.

In general, nanofluids could fall into the colloidal dispersion category, for a colloidal dispersion can be defined as a disperse phase in a dispersion medium [29]. Theoretical viscosity studies of colloidal dispersions were well documented by Goodwin and Hughes [30]. The investigation of the viscosity of dilute coarse colloidal suspensions was started by Einstein [31] by assuming a homogeneous distribution of rigid, uncharged spherical particles, and no attractive force between the particles. Also, under dilute conditions, it is assumed the perturbation around a particle is unbounded, which allows the hydrodynamic disturbances to decay to zero and therefore not affect any neighboring particles. In the liquid phase, there is an important assumption in his analysis, which is the surrounding fluid can be treated as a continuum. His approach yields the
effective viscosity of a colloidal suspension, $\mu_{\text{eff}}(\phi)$, as a function of particle
volume fraction $\phi$:

$$\mu_{\text{eff}}(\phi) = \mu_b(1 + 2.5\phi) \quad (1.1)$$

where $\mu_b$ is the viscosity coefficient of the base fluid. Note that eq. (1.1) is
only valid for $\phi<0.01$ [30]. Einstein’s viscosity model, however,
underestimates the effective viscosity of nanofluids even at very low
concentration [29, 32]. Moreover, Pak and Cho’s results [32] indicated
that the viscosity for $\gamma$-Al$_2$O$_3$ and TiO$_2$ nanofluids showed pseudoplastic
behavior at or above volume fractions of 3% and 10%, respectively.

1.3. Investigations of nanofluids convective heat transfer

Recently, there has been more attention paid to the convective heat
transfer performance of nanofluids [33-56], due to the recognition that in
practical applications nanofluids are likely to be used as heat transfer
fluids in flowing systems, such as microchannel or minichannel heat sinks.
However, the nanofluid convective heat transfer coefficient ($h$) or Nusselt
number ($Nu$) are inconsistent throughout the literature. For example, for
turbulent flow Xuan and Li [33] demonstrated that $Nu$ increased when the
volume fraction ($\phi$) of nanofluid and/or the Reynolds number, $Re$,
increased. In particular, the ratio of $Nu$ of copper-water (Cu-H$_2$O)
nanofluid to that of water varied from 1.06 to 1.39, while the volume
fraction of copper nanoparticles increased from 0.5% to 2.0%, at the same
constant $Re$. However, the results also illustrated that the Dittus-Boelter
correlation [57] failed to predict $Nu$ for nanoparticle $\phi$ greater then 0.5%.
Per the authors’ discussion, the enhanced energy exchange rate may be partially augmented by the random movement of nanoparticles. The particle random movements give rise to a slip velocity between the fluid and the particles and increase the temperature gradient between the fluid and the tube wall. Note that, from their pressure drop experiment, the nanofluid friction factors behave like their base fluid (water). Several groups [40, 43, 48 and 53] also tested nanofluids in the turbulent flow region with different nanoparticle material, base fluid, fluid mixtures and flow rate combinations. That body of research concluded that, with suspended nanoparticles, $h$ increases with increasing $Re$ and $\phi$ for turbulent flow. However, the enhancement magnitudes were not consistent.

Two groups [32, 44] observed controversial data of convective heat transfer performance in the turbulent flow region. Pak and Cho [32] conducted convective heat transfer experiments for $\gamma$-Al$_2$O$_3$ and titania (TiO$_2$) nanoparticles in water with $Re$ varying from $10^4$ to $10^5$. They observed a negative $h$ trend, such that $h$ was 12% lower compared with pure water at the same $Re$, for the nanofluid having a volume fraction of 3%. They believe the decrease was due to the significant increase in $\mu$ due to the addition of nanoparticles. Williams et al. [44] investigated Al$_2$O$_3$ and zirconia (ZrO$_2$) nanofluids at various flow rates ($9,000 < Re < 63,000$). They observed no abnormal heat transfer enhancement and hence the $Nu$
could be predicted by the traditional Dittus-Boelter model as long as the effective nanofluid properties are provided.

For nanofluids in a different test-section geometry, Sharma et al. [51] compared the nanofluid convective heat transfer performance in a circular tube with or without a twisted tape insert. Compared with water, the heat transfer enhancement for 0.1 vol% Al₂O₃-water nanofluid in [51] was 44.71% greater with twisted tape insert and 23.69% greater for a smooth circular tube at $Re=9,000$. In the research of Pantzali et al. [52], plate heat exchangers (PHE) were used and their results suggested nanofluids should be applied under laminar conditions. Nanofluid convective heat transfer in microchannels was also studied [38, 41 and 46]. Lee and Mudawar [38] found that overall cooling effectiveness is very small in their microchannels. Chein et al. [41] showed that a nanofluid can absorb more energy than its base fluid. However, Chein et al. also observed that when the flow rate is high, both nanofluid and its base fluid absorbed the same amount of energy. Jung et al. [46] studied a nanofluid containing 170-nm Al₂O₃ particles and observed a 32% increment in $h$.

1.4. Research objectives

From previous studies, it seems that with different particle/base fluid formula, particle shape or aspect ratio, test-section geometry and different $Re$ range, $h$ will have different increment magnitudes. Controversial results can be found in the literature. For example, data from Nguyen et al. [40] indicate smaller particles provide better heat
transfer while Kulkarni et al. [43] recommends larger diameter nanoparticles. Additionally, nanofluid convective research to date has focused on water-based nanofluids and at rather high fluid velocity. However, with the miniaturization trend of modern science and technology, attention should be paid to the laminar flow region to avoid extremely high pressure loss and viscous dissipation that may occur in small flow loops. Thus, in order to utilize nanofluids in conventional cooling applications, it is imperative to investigate convective heat transfer performance of nanofluids in the laminar flow region.

The specific questions being addressed in this dissertation are as follows:

1. How will nanoparticle loading affect laminar convective heat transfer and viscosity?
2. How will the physical properties of fluid mixtures affect laminar convective heat transfer and viscosity?
3. How does applied heat flux affect the nanofluid viscosity?
4. Are nanofluids better coolants than their base fluids?

This work focuses on experimental studies of viscosity and convective heat transfer of nanofluids. A general introduction has already been given above in Chapter 1. The experimental methodology and data analysis used for nanofluids viscosity and convection experiments are provided in Chapter 2. Since the thermophysical properties could be different if the nanofluid characteristics are different, the nanofluid preparation and characterization are presented in detail in Chapter 3. Chapter 4 and
Chapter 5 comprise the detailed literature review then provide the nanofluid viscosity and convective heat transfer empirical results in dimensional form, respectively. The nondimensional form of the convective heat transfer results is presented in Chapter 6. Finally, a summary and suggested future works are proposed in Chapter 7.
Chapter 2: Experimental methodology and apparatus validation of nanofluid viscosity and convective heat transfer coefficient

Measuring the convective heat transfer of nanofluids in a pipe is similar to measurements on other liquids. Nanofluids can be made stable against sedimentation through appropriate addition of surfactants [34], or through the addition of acids or bases [7] that increase the nanoparticle surface charge. Thus, each of these help to keep the nanoparticles separated from one another and in suspension. These additional chemicals, however, complicate the already complex nature of nanofluids, and render the analysis of the experimental results more difficult. In this research, instead of intentionally keeping the nanofluids stable for a long period, no alteration to nanoparticle surface, surfactants or any other chemicals was added to the tested nanofluids. Even without a stabilization agent, the nanofluids did not show significant sedimentation during the experiments. In this chapter, first the experimental methods used for measuring nanofluid viscosity and heat-transfer coefficient, and then system validation, are presented.

2.1. Experimental system for measuring nanofluid viscosity and convective heat transfer coefficient

An experimental setup, shown as Fig. 2.1, was designed and assembled to permit systematic measurements and analysis of nanofluid viscosity and heat transfer coefficient. The experimental system included
Fig. 2.1. Schematic diagram of experimental apparatus for convective heat transfer and viscosity measurement.

Fig. 2.2. Sketch of heater wire and thermocouple locations (not drawn to scale).

Fig. 2.3. Cross-sectional view of test tube (not drawn to scale).
one sample tank, a calibration tank, a pump, a gauge pressure transducer, a differential pressure transducer, a heating jacket, a cooling jacket, connecting piping, and a single 1.02-mm inner diameter and 50-cm-long straight stainless steel tube (AISI 316) served as the test section. The heating and cooling jackets were two simple counter-flow heat exchangers which keep a constant bulk temperature at the inlet of the test section. The test tube was heavily insulated because it was wrapped in a fiberglass blanket (thermal conductivity, $k$, of 0.03 Wm$^{-1}$K$^{-1}$) then sandwiched between two 2-in.-thick calcium silicate boards ($k = 0.1$ Wm$^{-1}$K$^{-1}$). The portions upstream and downstream of the test section were another two 10-cm long stainless steel tubes, which had the same inner diameter and were insulated in the same way as the test section. The connections between these 10-cm long tubes and the test section also had an inner diameter of 1.02 mm. The test fluids were driven by a low pressure, micro annular gear pump (Micropump Inc, Model No. 2521) over a volume flow rate capacity of 0.15 – 9 ml/min. The volume flow rates during the experiments were controlled by a computer through a controller provided by Micropump Inc. The pressure difference across the hydraulically fully developed flow test section was measured by a differential pressure transducer (Setra Inc., Model 230). Based on the flow rates during the experiments, the 10-cm-long upstream section was sufficient to produce hydraulically fully developed flow at the entrance of the test section. Therefore, pressure drop data gathered during convective heat transfer
experiments were sufficient to calculate the fluid viscosity via Poiseuille’s law [57] and tests of heat transfer coefficient could be carried out in both the thermally developing flow region, and in the thermally fully-developed flow region. The pressure drop measurements were conducted before and during heat transfer measurements to explore the temperature effect on the hydrodynamic behavior of nanofluids.

The test tube is constantly heated by a polyimide-coated nichrome wire (Lakeshore NC-32) that is wrapped in a non-inductive fashion around the outside tube wall. The non-inductive wrapping is done by doubling the heater wire and then winding the test tube like a solenoid (Fig. 2.2). Therefore, at every point along the wire there are equivalent currents passing in both directions, thus roughly canceling out any induced magnetic field. The input power was confirmed by a digital multimeter. Two T-type thermocouples were inserted into the flow field to measure the nanofluid’s mean entrance and exit temperatures. Another 12 T-type thermocouples were mounted on the test tube wall, by high-thermal-conductivity epoxy, as sketched in Fig. 2.2. The staggered separation of the thermocouple locations was used to carefully monitor the temperature profile in the entrance region. All temperature and pressure data were recorded into a computer through a data acquisition system from National Instruments.
2.2. Data analysis for nanofluid viscosity

With respect to viscosity, the suspensions with spherical or nearly spherical solid particles differ from polymer colloids. At low volume fraction, the viscosity of the former suspensions does not usually depend on the shear stress or on the method of measurements [58]. The particle size and shape characterization will be presented later on and those images indeed show spherical or nearly spherical nanoparticles. Therefore, without surfactant, which is usually long-chain polymers, it is rational to acquire the effective viscosity of nanofluid via data from differential pressure measurements and Poiseuille’s law [57].

Due to the limitations of the annular gear pump, the maximum $Re$ did not exceed more than 400 in all cases. When $Re < 2300$, the flow regime is categorized as the laminar flow region and the pressure drop ($\Delta P$) can be expressed as [57]

$$
\Delta P = \frac{32 \mu U_m L}{D_i^2}
$$

(2.1)

where $L$ is the distance between the two pressure taps, $\mu$ the effective viscosity of the test fluid, $D_i$ the inner diameter of the test tube, and $U_m$ the mean flow velocity which can be related to volume flow rate ($\dot{V}$).

Therefore, with known $\dot{V}$ and the measured $\Delta P$ across the test section, one can calculate the effective viscosity of the test fluid by eq. (2.2), which is a form of Poiseuille’s law:
\[ \mu = \frac{\pi D^4 \Delta P}{128 L V} \]  

(2.2)

2.3. Data analysis for nanofluid convective heat transfer coefficient

Owing to the simple test-tube geometry, once the surface heat flux \((q'')\), local inner-wall temperature \((T'_w(x))\), and local fluid temperature \((T_f(x))\) are obtained, the local heat transfer coefficient, \(h(x)\), can be evaluated by Newton’s law of cooling [57]:

\[
h(x) = \frac{q''}{T'_w(x) - T_f(x)}
\]  

(2.3)

where \(q''\) is generated by the Nichrome heater wire and calculated by Joule’s law:

\[
q'' = \frac{Q}{A_h} = \frac{I \cdot V}{2 \pi r_i L}
\]  

(2.4)

where \(Q\) is the supplied heat rate which is directly related to \(I\) the measured current and \(V\) the supplied voltage as in eq. (2.4), \(r_i\) the inner radius of the test tube (as shown in the cross-sectional view of the tube of Fig. 2.3) and \(L\) the heating length.

\(T'_w(x)\) is the inner wall temperature calculated from the steady one-dimensional heat conduction equation in cylindrical coordinates [57], i.e.,

\[
\frac{1}{r} \frac{d}{dr} \left( r \frac{dT}{dr} \right) = 0
\]  

(2.5)

with constant heat flux boundary condition at the wall, hence the solution of \(T'_w(x)\) becomes

\[
T'_w(x) = T_s(x) - \frac{Q \cdot \ln(r_e/r_i)}{2 \pi L k_i}
\]  

(2.6)
where \( T_{w}(x) \) are the local outer wall temperatures, which were measured by T-type thermocouples mounted on the tube wall. \( r_o \) is the outer tube radius \( r \), and \( k_t \) is the thermal conductivity of the tube wall. Typical calculated temperature differences between the inner and outer walls ranged from 0.02 to 0.05 °C. The local fluid temperature, \( T_f(x) \), is determined by an energy balance:

\[
T_f(x) = T_{f1} + \frac{Q}{\rho \dot{V}} \frac{x}{L}
\]

(2.7)

where \( T_{f1} \) is the fluid temperature at the inlet, \( \rho \) the nanofluid density, \( c \) the nanofluid heat capacity, and \( \dot{V} \) the fluid volumetric flow rate. \( x \) is the axial distance from the entrance of the test section. The nanofluid density is calculated by \( \rho = \varphi \rho_p + (1 - \varphi)\rho_b \). As for \( c \), measurements from Zhou et. al. [59] suggest the following formula:

\[
c = \frac{\varphi \rho_p c_p + (1 - \varphi)\rho_b c_b}{\rho}
\]

(2.8)

2.4. Calibration and uncertainty analysis

The microannular gear pump can maintain the accuracy of volume flow rate to ±0.5% and was provided by Micropump Inc. The differential pressure transducer which has ±0.25% accuracy was calibrated by its manufacturer, Setra. The T-type thermocouples were calibrated within a thermostat-controlled distilled-water bath and the accuracy can reach ±0.1%. The accuracy for the pH meter is ±0.02 and ±0.5% for the electrical conductivity meter. Extraneous losses from the heater were estimated to be less than 3%, based on measurements for pure water.
Each measured point on the results graphs represents the average of the values obtained from three experimental runs. The uncertainty of experimental data was estimated by the methods described by Figliola and Beasley [60] and was calculated based on a 95% confidence level. Student t-distribution was used during the precision error calculation for the sample size of three.

2.5. Apparatus validation

Using the aforementioned experimental procedures and analysis methods, the experimental equipment was validated with pure water by measuring the pressure drop ($\Delta P$) and $h$ across the test section. The pressure drop results for pure water are provided in Fig. 2.4. The measured data points agreed within 3% of the theoretical line; therefore, the assumption of a steady laminar flow with hydrodynamically fully developed condition is valid. The theoretical line for pure water is calculated by eq. (2.1).

In the fully developed region with constant surface heat flux, $Nu$ has an analytical solution and is equal to a constant $Nu=4.364$ [61] for round tubes. The Kays and Crawford model [61] for $Nu(x)$ in the developing region is frequently
Fig. 2.4. Pressure drop across the test section using pure DI-H$_2$O (no nanoparticles) as the working fluid.

Fig. 2.5 Comparison between measured and calculated [61] heat transfer coefficients for developing flow, for pure DI-H$_2$O (no nanoparticles)
cited by many research groups for laminar flow with the constant heat flux boundary condition. The theoretical solution for the local $Nu$ number in the thermal entrance region in [61] is expressed as

$$Nu(x) = \left[ \frac{1}{Nu_\infty} - \frac{1}{2} \sum_{m=1}^{\infty} \frac{\exp(-\gamma_m^2 x^+)}{A_m \gamma_m^4} \right]^{-1}$$

(2.9)

where $Nu_\infty=4.364, x^+=\frac{2x}{D_{RePr}}$, $\gamma_m$ and $A_m$ are corresponding eigenvalues and constants, which are provided in Table 9-5 in [61]. Here, for comparison with raw data of nanofluids, we calculate $h(x)$ from $Nu(x)$:

$$h(x) = \frac{Nu(x) k_{H_2O}}{D_i}$$

(2.10)

Figure 2.5 shows the local $h$ results of pure DI- H$_2$O, in both the thermally developing region and the fully developed region, together with the theoretical solution from the Kays and Crawford’s model [61] within the laminar flow regime under constant heat flux boundary condition. In the fully developed region, the calculated and measured $h$ values both vary because of the temperature dependence of $k_{H_2O}$. The lower the volume flow rate is, the higher the water temperature becomes under the same heat flux. Hence, the higher the water temperature, the higher $k_{H_2O}$ will be which results in higher $h$ at the same $Nu$. The results show that the measured pure water $h$ values for the test section are consistent with the predictions. Furthermore, the experimental data exhibit good agreement with the trend of the classical thermal-entry length relation [61]:
\[
\left( \frac{x}{D_t} \right)_{fully\ developed} = 0.05 \cdot \text{Re} \cdot \text{Pr} \tag{2.11}
\]

2.6. Summary

The experimental instrument and data analysis method of viscosity and convective heat transfer performance of nanofluids was introduced in this chapter. The test section was heavily insulated and had a designated hydrodynamic developing region adjacent to the test tube. The system validation was done by pure DI-H_2O and the results were presented in section 2.5. The system validation shows good agreement with both the pressure drop and convection heat transfer theoretical predictions.
Chapter 3: Nanofluid preparation, characterization and stability.

As pointed out by some reviewers [18, 23, 25], attention to nanofluid preparation and several characterization parameters is very important for the research groups working with nanofluids. The nanoparticle synthesis method and the nanofluid preparation method can affect the nanofluid thermophysical properties because the surface chemistry of the nanoparticles is changed. In consequence, this alteration in nanoparticle surface chemistry can also affect nanofluid stability. The characterization parameters such as the size and shape of nanoparticles or agglomerates, pH, electrical conductivity ($\sigma$) and zeta potential ($\zeta$) within the nanofluids not only supply information on the nanofluid aggregation stage and stability, but may also provide insights into certain enhancement mechanisms of the nanofluid. Take $k$ as an example; it has been presented that particle agglomerates play an essential role in the magnitude of enhanced $k$ of nanofluids [62, 63]. Recently, Lee et al. [64] experimentally investigated the influence of the surface chemical effect on $k$. Their results point out that a higher surface charge is desired. Therefore, in this chapter, nanoparticle and nanofluid characterizations will be presented after a description of the nanofluid preparation process. After the characterization data, one will be able to calculate the nanofluid stability ratio. Hence, a short discussion in regards to nanofluid stability will also be provided.
3.1. Nanofluid preparation.

There are two major ways to disperse nanoparticles into base fluids. One is the VEROS method [65], also known as the “one-step” method, which allows simultaneously producing and dispersing nanoparticles into base fluids. The other is the so-called “two-step” method. In the second method, the nanoparticles are dispersed in a base fluid by ultrasonification. The two-step technique was chosen to be the nanofluids preparation method because the method worked well for oxide nanopowders [18, 21] and lots of different oxide nanopowders can be purchased nowadays.

The nanoparticles were purchased from Nanostructured & Amorphous Materials, Inc, and were made by a plasma process from conductive material. Although many different types of nanofluids have been tested in various labs around the world, the alumina/water nanofluids are meant to be representative of this large (and growing) body of nanofluids data. Therefore, the main test samples were γ-Al₂O₃ nanoparticles in DI- H₂O and propylene glycol (PG) in DI- H₂O mixtures. PG is an organic compound which is clear, odorless, viscous and totally miscible with water. PG has the chemical formula of C₃H₈O₂. Because of its low freezing point (below -60°F), PG is commonly used in antifreeze solutions and heat transfer media [67]. The volume ratios of PG: DI- H₂O were 9:95 and 15:85 and will be reported as 5/95 PG/H₂O and 15/85 PG/H₂O respectively. Table 3.1 shows some thermal properties of all base fluids at 25 °C. The γ-Al₂O₃ nanoparticles have 99.97% purity, a mean
Table 3.1

Properties of base fluids at 25 °C.

<table>
<thead>
<tr>
<th></th>
<th>Density [kg m⁻³]</th>
<th>Viscosity [mPa s]</th>
<th>Thermal conductivity [W m⁻¹ K⁻¹]</th>
<th>Specific heat [J Kg⁻¹ K⁻¹]</th>
</tr>
</thead>
<tbody>
<tr>
<td>DI-H₂Oᵢ</td>
<td>997.0</td>
<td>0.891</td>
<td>0.607</td>
<td>4180</td>
</tr>
<tr>
<td>PGᵢ</td>
<td>1036.1</td>
<td>60.5</td>
<td>-</td>
<td>2483</td>
</tr>
<tr>
<td>5/95 PG/H₂Oᵢ</td>
<td>1001.1</td>
<td>0.945</td>
<td>0.576</td>
<td>4133</td>
</tr>
<tr>
<td>15/85 PG/H₂Oᵢ</td>
<td>1011.7</td>
<td>1.495</td>
<td>0.524</td>
<td>4033</td>
</tr>
</tbody>
</table>

diameter of 20 nm, specific surface area of 180m²g⁻¹ and density of 3900 kg m⁻³ as reported by the manufacturer.

Starting from a desired volume fraction value, the corresponding mass of γ-Al₂O₃ nanoparticles was calculated and weighed. Second, the corresponding volume of base fluid was measured. Because the nanoparticle manufacturing process could result in a large degree of agglomeration, the nanoparticles were finally dispersed into the base fluid via ultrasonication. The ultrasonication (model UP200S from Dr. Hielscher GmbH) was carried out for 1 hour to break up any potential clusters in the dry nanoparticle powder with a power of 40W during ultrasonication process. After ultrasonication, the nanofluids were rested at room temperature for one hour to eliminate any heating effects from the ultrasonication. The volume fractions of the test samples were 0.5%,

¹ Data are from [68].
² Data are from [67].
0.75% and 1%. Again, there was no surfactant used in any of the samples reported.

3.2. Size and shape of dry nanoparticles

Since most other properties are affected to a certain extent by the particle size and shape, these two factors are the most decisive characteristics of nanofluids [29]. For size measurement down to a few nanometers, researchers are likely to use high-resolution electron microscopes, especially transmission electron microscopes (TEM), because TEMs are able to resolve down to 0.2nm. The operating principle of TEM shares the same idea as a light microscope, but shines a beam of electrons instead of light. TEM has the advantage of developing real space images for direct observation. The TEM images of dry nanoparticles are presented as Fig. 3.1, which shows an overall view of clusters ranging from ~100 nm to a few μm. Since the size of the particles was not uniform, there were aggregations in the tested dry nanopowder. The bright gray ovals are sample-holding substrate. The dark black spots are aggregates of dry nanoparticles.
Fig. 3.1. Transmission electron microscope (TEM) image for 20-nm dry 
Al$_2$O$_3$ nanopowder. The clusters show size ranges from hundred nm to a 
few μm. The bright gray oval spots are sample-holding substrate. The 
dark black spots are due to strong agglomeration of dry nanoparticles.

These sample nanoparticles were never dispersed into liquid, nor went 
through the ultrasonication process. Fig. 3.2 (a) is another TEM image of 
dry nanopowder, which gives an idea of the shapes of nanoparticles and 
clusters at a closer view. The image shows that most nanoparticles have a 
spherical or nearly spherical shape and partially arbitrary shapes. When 
observed more closely most single alumina crystals have sizes ranging 
from 10-20 nm. In Fig. 3.2 (b), the sample was first prepared as 1 vol% 
Al$_2$O$_3$-H$_2$O nanofluids and ultrasonicated for an hour, like the 
aforementioned sample-preparation procedures; then, a small amount
was transferred to a substrate. The sample was spread in a very thin layer, and allowed to dry out on the supporting substrate. The image again shows that there are aggregates, but does not show a thick pile-up that appears similar to the dry nanopowder. Both Fig. 3.2 (a) and (b) show the size of the nanoparticles is not homogenous, and that there is a size distribution of the nanoparticles. However, there is no direct evidence to tell if the aggregates are from the dry nanopowder, the agglomerations in the nanofluid, or particles piling up when waiting for water to evaporate. This is always a main drawback when applying TEM on wet samples. Extensive sample preparation before the sample is transferred into the vacuum chamber of the microscope may ruin the structure of the sample at the same time. Therefore, a non-invasive size measurement technique such as dynamic light scattering (DLS) is favored, as explained in the next section.
Fig. 3.2. (a) TEM image for dry Al$_2$O$_3$ nanopowder. The image shows clusters of spherical, nearly spherical and some arbitrary shaped nanoparticles.

Fig. 3.2. (b) TEM image for 1 vol% Al$_2$O$_3$-water after water is evaporated.
3.3. Nanofluid characterization

3.3.1. Nanoparticle size in suspension

The idea of the DLS technique depends on the random nature of nanoparticles rotating, translating, and vibrating in base fluids, and, once exposed by a light source, these particles move and cause randomly scattered light fluctuations in the illuminated region. Embedded in these fluctuations is the dynamical and structural information of the particle position and orientation. The particle size distribution within the fluid can be obtained by relating the scattered light signals of random fluctuations to the diffusion coefficient of nanoparticles, which is the well-known Stokes-Einstein relation, via the decay time of the autocorrelation function of the scattered light intensities, assuming the particles or clusters are spherical in shape [66]. The autocorrelation function expresses the degree of similarity between scattered intensities with respect to time. For the detailed theory and calculation of the autocorrelation function, please refer to Berne and Pecora [66]. The DLS instrument used in this study is Nicomp 380/ZLS from Particle Sizing Systems, Inc. The system is equipped with a 50mW HeNe laser and is capable of measuring particle sizes ranging from 1nm to 5μm. The laser wavelength is 639nm and the measured angle is set at 90.0 degrees which is the default setting from manufacturer.
3.3.2. Nanofluid pH, electrical conductivity and zeta potential

When dispersing Al₂O₃ nanoparticles into any base fluid, the particle surface can acquire an electric charge by absorbing or desorbing protons at the particle/liquid interface, especially when the base fluid is a polar medium like DI-H₂O [29]. This absorbing and desorbing mechanism forms two layers that surround the particle surface. The inner region is the Stern layer, where the ions are strongly attached to the particle surface. The diffuse layer, which is the outer layer, contains ions that are not firmly bound. The potential at this electrical double layer (EDL) boundary is known as the zeta potential (ζ). The magnitude of ζ represents the strength of the electrostatic energy barrier between particles. A greater ζ increases the interparticle repulsion in a nanofluid of similar nanoparticles. Hence, less aggregation will occur and the nanofluids will be more stable. The ζ and the thickness of the EDL are strongly dependent on the pH value. Once the pH value exceeds a certain limit, the ions cause significant shrinkage of the EDL, and the nanofluid is no longer be stable. The pH values were measured by a pH meter (Hach EC10). In the presence of the EDL, the σ of a suspension can be increased due to the surface conductivity of the particles in the EDL region. The nanofluids’ σ values were measured by a Hach CO150 meter. The pH meter was calibrated using a single point calibration technique, with a Hach standard buffer solution of pH 7.00±0.02. The electrical
conductivity meter was calibrated using a single point calibration technique with a standard buffer.

The same DLS instrument, Nicomp 380/ZLS, can also perform electrophoretic light scattering (ELS) and measure $\zeta$ in the sub-micron range. The ELS technique is based on measuring the light scattered from moving particles under the influence of alternating electric fields. The difference between DLS and ELS measurements is the subjected particle motion. In DLS, the Brownian motion causes the frequency or phase shift of the incident laser beam(s). In the case of ELS, the oscillating electric field performs the same function, and the particle velocity is determined by a superposed spectrum shift of Brownian diffusion, plus the particle velocity caused by the electric field. If an electric field is applied to the nanofluid in a DLS system, the particles will move toward the particular direction in response to the field. Once the effect of the Brownian motion is eliminated, the particle velocity caused by the electric field is directly proportional to the magnitude of the electric field and the proportionality constant is defined as electrophoretic mobility ($\eta$). The $\eta$ can be computed from the measured frequency. Once $\eta$ is known, then $\zeta$ is related to $\eta$ through $\zeta = \mu \eta / \epsilon$, known as the Smoluchowski formula [29], where $\mu$ is the viscosity and $\epsilon$ is the dielectric constant of the base fluid.

Because DLS and ELS results are strongly dependent on the data of the light scattering correlation, once the nanofluid gets too turbid the laser either cannot pass through the sample or causes erroneous results from
multiple scattering. To eliminate multiple scattering, diluted samples are required. The appropriate dilution was determined by the power spectrum (shown as Fig. 3.3) during the ζ measurements. The sharp spectrums indicate sufficient scattered light and can provide adequate data for analysis. The diluted sample concentration for both the size and ζ measurements was 0.0006 vol%. In other words, three dispersions with volume fractions of 0.5%, 0.75% and 1% were first prepared, and then all three samples were diluted to 0.0006 %. After dilution, the zeta potential measurement was carried out before the size measurement.

![Power Spectrum](image)

**Fig. 3.3.** Example of a good power spectrum during zeta potential measurements for determining nanofluids concentrations for DLS measurement. The red and black curves are the reference and the sample relative frequencies.
3.3.3. Results of nanofluid characterization

The pH and σ values for the original and the diluted samples were measured and are summarized in Table 3.2. The “Volume Fraction (%)” column indicates the volume fractions of the undiluted samples (i.e., the “original” samples). The “(original)” columns represent that the results were measured with undiluted samples (i.e., 0.5, 0.75 and 1 vol% nanofluids) and the “(diluted)” columns represent that the diluted samples were prepared from the original samples and had a volume fraction as 0.0006 vol%. The DI- H2O for water-based nanofluids has a pH of 7.40 and σ of 1.93μS/cm. For nanofluids using PG/H2O mixtures as base fluids, the DI- H2O has a pH of 7.36 and σ of 2.03μS/cm because the water was harvested at a different time from the same machine. The PG has a pH of 8.04 and σ of 0μS/cm. As shown in Table 3.2, the original undiluted samples had pH values greater than 8 no matter in which base fluid. The results of DI-H2O based nanofluids show that with an increasing volume fraction, the pH has a peak at 0.75% for the original undiluted samples. However, for the diluted samples, the pH has a minimum at 0.5 % and other samples were all close to 7.4-the base fluid pH. For PG/H2O based nanofluids, the pH increases with increasing particle volume fraction for all undiluted samples which has a maximum value at 1 vol% Al2O3-15/85 PG/H2O nanofluid sample and this increasing trend is similar to [68, 69]. However, for the diluted samples, the 5/95 PG/H2O samples have pH values around 7.6 but 15/85 PG/H2O samples have an enormous increase
at 1%. The $\sigma$ data shows an increasing trend with increasing particle loading, which is consistent with [70] for all original undiluted samples. The $\sigma$ data of DI-H$_2$O based nanofluid are much higher than PG/H$_2$O-mixture ones and have a higher rate of increase with respect to nanoparticle volume fraction. This could be due to two possible reasons: first, the contribution of zero electrical conductivity of PG and second, the PG modifies the nanoparticles’ surface chemistry. However, for diluted samples, the $\sigma$ data did not show a conclusive trend.
Table 3.2

pH and electrical conductivity of alumina nanoparticles in different base fluids.

<table>
<thead>
<tr>
<th>Volume Fraction</th>
<th>pH (original)</th>
<th>σ [μS/cm] (original)</th>
<th>pH (diluted)</th>
<th>σ [μS/cm] (diluted)</th>
</tr>
</thead>
<tbody>
<tr>
<td>DI-H₂O</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Base fluid</td>
<td>7.40 ± 0.03</td>
<td>16.9 ± 0.06</td>
<td>7.40 ± 0.03</td>
<td>16.9 ± 0.06</td>
</tr>
<tr>
<td>0.5 %</td>
<td>8.24 ± 0.05</td>
<td>270.00 ± 6.57</td>
<td>7.18 ± 0.05</td>
<td>623.33 ± 1.43</td>
</tr>
<tr>
<td>0.75 %</td>
<td>8.44 ± 0.06</td>
<td>376.67 ± 28.25</td>
<td>7.45 ± 0.23</td>
<td>146.33 ± 0.15</td>
</tr>
<tr>
<td>1 %</td>
<td>8.37 ± 0.05</td>
<td>448.00 ± 49.69</td>
<td>7.47 ± 0.14</td>
<td>108.87 ± 0.13</td>
</tr>
<tr>
<td>5/95 PG/H₂O</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Base fluid</td>
<td>7.45 ± 0.04</td>
<td>1.78 ± 0.07</td>
<td>7.45 ± 0.04</td>
<td>1.78 ± 0.07</td>
</tr>
<tr>
<td>0.5 %</td>
<td>8.31 ± 0.06</td>
<td>13.20 ± 0.52</td>
<td>7.62 ± 0.05</td>
<td>10.20 ± 0.41</td>
</tr>
<tr>
<td>0.75 %</td>
<td>8.43 ± 0.06</td>
<td>20.12 ± 0.73</td>
<td>7.55 ± 0.05</td>
<td>9.4 ± 0.35</td>
</tr>
<tr>
<td>1 %</td>
<td>8.49 ± 0.05</td>
<td>29.6 ± 0.89</td>
<td>7.58 ± 0.04</td>
<td>10.6 ± 0.42</td>
</tr>
<tr>
<td>15/85 PG/H₂O</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Base fluid</td>
<td>7.51 ± 0.03</td>
<td>3.14 ± 0.10</td>
<td>7.51 ± 0.03</td>
<td>3.14 ± 0.10</td>
</tr>
<tr>
<td>0.5 %</td>
<td>8.52 ± 0.05</td>
<td>7.94 ± 0.13</td>
<td>8.01 ± 0.04</td>
<td>6.29 ± 0.18</td>
</tr>
<tr>
<td>0.75 %</td>
<td>8.65 ± 0.06</td>
<td>9.62 ± 0.11</td>
<td>8.03 ± 0.06</td>
<td>6.32 ± 0.21</td>
</tr>
<tr>
<td>1 %</td>
<td>8.83 ± 0.06</td>
<td>15.09 ± 0.21</td>
<td>8.36 ± 0.11</td>
<td>10.09 ± 0.23</td>
</tr>
</tbody>
</table>
Fig. 3.4. Zeta potential vs time after sonication for diluted Al₂O₃ nanofluids at different original (undiluted) volume fractions.

The $\zeta$ as a function of time after sonication is shown in Fig. 3.4 for the diluted samples. The results show two distinct groups. The DI-H₂O based nanofluids are in the higher value group which has $\zeta \sim 47$ mV after 30 mins. It is also shown that the $\zeta$ for the DI-H₂O based nanofluids with 0.75% and 1% volume fractions are at maximum at 20 mins and then decrease, while the zeta potential for the 0.5% samples remains almost constant around 47mV. For the $\zeta$ of PG/H₂O based nanofluids, results for 5/95 PG/H₂O are limited in a gap between 30-33mV while the 15/85 PG/H₂O based nanofluids have $\zeta$ falling below 30mV. All positive $\zeta$
suggests that all nanoparticles acquired positive surface charge in the
nanofluids in this study. The significant drop in \( \zeta \) of 1 vol\% \( \text{Al}_2\text{O}_3 \)-15/85
PG/H\(_2\)O sample is expected because the sample has a pH value at 8.36
which is closed to the pH value of the isoelectric point (IEP) of alumina
colloidal solution. The term IEP is the pH value which represents when
the net charge at the immersed particle surface is zero and thus \( \zeta \) equals
zero. The distribution of IEP published for alumina is nearly Gaussian
with a peak at pH 8.8 as collected in Kosmulski’s literature survey over the
period 1966-1999 [71].

In general, nanoparticle dispersions have a tendency to aggregate
due to attractive forces between the particles. Therefore, a given
dispersion is likely to exhibit several size distributions over time as a result
of different aggregation rates. The Nicomp 380/ZLS is equipped with
software which allows us to analyze bimodal populations, and an example
of which is shown in Fig. 3.5 for one of our samples. Figure 3.5 is a
volume-weighted distribution, not an intensity-weighted result.
Recognizing that larger particles generally scatter more light than smaller
particles, using an intensity-weighted distribution may lead to an opposite
interpretation of the particle size distribution.
Fig. 3.5. An example of a particle size distribution measured with the Nicomp 380/ZLS system. Screen shot is the particle sizing result of diluted 20-nm $\gamma$-Al$_2$O$_3$ nanoparticles in DI-H$_2$O taken 70 minutes after sonication. The distribution is generated by the Nicomp volume-weighted model.

The evolutions of the average aggregate size over time, in diluted samples, are presented in Figs. 3.6-3.8. The arrangement of the figures is as follows: Figs. 3.6 (a)-(c) are data for DI-H$_2$O based nanofluids, Figs. 3.7 (a)-(c) are results for 5/95 PG/H$_2$O based nanofluids and finally Figs. 3.8 (a)-(c) are for 15/85 PG/H$_2$O based nanofluids. The volume % for each aggregate size population is indicated on the y-axis in Figs. 3.6-3.8. This
information provides the percentage of aggregate size in each tested nanofluids. Taking Fig. 3.6 (a) as an example, the results show there was around 51.5% nanoparticles in the size range of 100.7±15.4nm and 48.5% nanoparticles were in the size range of 360±46.8nm. All three (a) figures give the results taken 60 minutes after sonication, (b) figures give the results 10 minutes after (a), and the (c) data were taken after another 10 minutes.

Fig. 3.6. (a) Diluted aggregate mean diameters and % of volume vs. original nanoparticle volume fraction 60 mins after sonication (DI-H₂O).
Fig. 3.6. (b) Diluted aggregate mean diameters and % of volume vs. original nanoparticle volume fraction 70 mins after sonication (DI-H₂O).

Fig. 3.6. (c) Diluted aggregate mean diameters and % of volume vs. original nanoparticle volume fraction 80 mins after sonication (DI-H₂O).
In Figs. 3.6 (a)-(c), it appears that the 0.5% nanofluid, in particular, manifests a stunning size fluctuation over the measured 30-minute time interval. The explanation for this observation may be due to the following issues. First, the dry nanoparticles have arbitrary shapes as shown in Fig. 3.2. Because the DLS technique usually assumes the tested particles are spherical, the arbitrary shape could have an elongated axis and hence the DLS measurements will have different results once the particle orientation has changed. Second, the zeta potential—around 47 mV—only provides partially repulsive force to avoid aggregation or weak flocculation. Thus, one may see mean aggregate diameters of 100.7 nm and 360 nm initially, and then see three peaks 20 mins later. Finally, it might be due to the suspiciously high $\sigma$ as shown in Table 3.2. Besides 0.5% nanofluid, it appears that the size and percent volume of particle size population of the other nanofluids remains steady throughout the measurements, which indicates that the 0.75% and 1% nanofluids are relatively stable compared with the 0.5% ones.

For DLS size measurements of PG/H$_2$O based nanofluids presented as in Fig. 3.7 (a)-(c) and Fig. 3.8 (a)-(c), it is shown that cluster size and aggregation rate increase with increasing nanofluid volume fraction. All PG/H$_2$O mixtures based nanofluids have greater cluster size and aggregation rate when compared with H$_2$O based nanofluids. Additionally, the % volume of particle size population of PG/H$_2$O based nanofluids increases with time in all cases, and especially more significant increases
were observed when the original nanofluid had a higher volume fraction. The cluster size and aggregation rate of 5/95 PG/H2O based nanofluids are less than those of 15/85 PG/H2O based nanofluids. These large aggregation rates and dramatic changes in the size distribution of PG/H2O based nanofluids could be due to the relatively low ζ value and from the pH value which is close to I.E.P.

So far, it is obvious that pure nanofluid characterization does not provide sufficient and conclusive information of nanofluid stability, particularly the cluster size from DLS reveals particles still undergoing aggregation and segregation when high ζ is observed. Therefore, a quantitative representation which determines if nanoparticles coagulate or remain separated in liquid will be presented in the next section.

Fig. 3.7. (a) Diluted aggregate mean diameters and %volume vs. original nanoparticle volume fraction 60 mins after sonication (5/95 PG/H2O).
Fig. 3.7. (b) Diluted aggregate mean diameters and %volume vs. original nanoparticle volume fraction 70 mins after sonication (5/95 PG/H₂O).

Fig. 3.7. (c) Diluted aggregate mean diameters and %volume vs. original nanoparticle volume fraction 80 mins after sonication (5/95 PG/H₂O).
Fig. 3.8. (a) Diluted aggregate mean diameters and %volume vs. original nanoparticle volume fraction 60 mins after sonication (15/85 PG/H₂O).

Fig. 3.8. (b) Diluted aggregate mean diameters and %volume vs. original nanoparticle volume fraction 70 mins after sonication (15/85 PG/H₂O).
3.4. Nanofluid stability.

3.4.1. Calculation of Nanofluid stability ratio.

Stability can be referred to as the nanoparticles distributed uniformly in suspension. Without surfactant, the electrostatic potential becomes the main mechanism to keep the particles separated and the ζ results in the previous section give qualitative information on nanofluid stability. The quantitative concept of nanofluid stability is the stability ratio ($W$) which measures the efficiency of the potential energy barrier and prevents particles from aggregation. To obtain $W$, one can start with the Derjaguin-Landau-Verwey-Overbeck theory (DLVO) which states the agglomeration of nanoparticles depends on the balance of attraction
potential energy (van der Waals interaction potential, $V_A$) and electrostatic repulsion potential energy ($V_R$) between particles. $V_R$ depends on the strength of the particle electrical barrier and is based on the results of $\zeta$ characterization. $V_A$ between two spheres is modelled by [29]

$$V_A(s) = -\frac{A_{131}}{6} \left( \frac{2r_p^2}{s(s + 4r_p)} + \frac{2r_p^2}{(s + 2r_p)^2} + \ln\left(\frac{s(s + 4r_p)}{(s + 2r_p)^2}\right) \right)$$

(3.1)

where $A_{131}$ is the Hamaker constant and $s$ the inter-particle distance. $V_R$ is given by [28, 70]

$$V_R(s) = 2\pi e_o \varepsilon_r \kappa^2 \varepsilon_p^2 e^{-\kappa s}$$

(3.2)

where $e_o$ is the dielectric permittivity of a vacuum, $\varepsilon_r$ the relative dielectric permittivity of the liquid and $\kappa$ the Debye-Hückel parameter. $\kappa^{-1}$ is the so-called Debye length. It is worth noting that eq. (3.2) is only valid for small particles or large Debye length which can be represented as $\kappa r_p < 5$. $\kappa$ depends on the temperature and the bulk electrolyte concentration.

Hence, $\kappa$ can be written as [29]

$$\kappa = \left(\frac{2000F^2 I_s}{e_o \varepsilon_r R_c T}\right)^{\frac{1}{2}}$$

(3.3)

where $F$ is Faraday’s constant, $R_c$ the universal gas constant and $I_s$ the ionic strength which can be related to $pH$ value when salt is absent.

$$I_s = 10^{-pH} \text{ for } pH \leq 7$$

(3.4)

$$I_s = 10^{pH-14} \text{ for } pH > 7$$

(3.5)
Therefore, $W$ can be calculated by the total interaction potential $V_T = V_A + V_R$ and written as [29]

$$W = 2r_p \int_0^\infty \frac{\exp\left(\frac{V_T}{k_BT}\right)}{(s+2r_p)^2} ds$$  \hspace{1cm} (3.6)

The lower limit of $W$ equals to unity and occurs when $V_T=0$, i.e. in the absence of repulsion. Overall, once $W$ is greater than $10^5$, the suspension is then considered stable [29]. In order to correct the hydrodynamic interaction on the particle coagulation, a correlation factor $B(s)$ was introduced by Honig [73] as follows:

$$B(s) = \frac{6\left(\frac{s}{r_p}\right)^2 + 13\left(\frac{s}{r_p}\right) + 2}{6\left(\frac{s}{r_p}\right)^2 + 4\left(\frac{s}{r_p}\right)}$$  \hspace{1cm} (3.7)

Therefore, the final form of $W$ is

$$W = 2r_p \int_0^\infty \frac{B(s)\exp\left(\frac{V_T}{k_BT}\right)}{(s+2r_p)^2} ds$$  \hspace{1cm} (3.8)

3.4.2. Results of nanofluid stability ratio.

By using eq. (3.1-3.8) and pH and $\zeta$ values from the characterization measurements, the nanofluid stability ratio results are presented in Table 3.3. The table also includes the Hamaker constant ($A_{131}$) of each case. The results show, as expected, that $W$ decreases with increasing particle loading within the same base fluid. Compared to different base fluids, it is
shown that 15/85 PG/H₂O based nanofluids were less stable compared with pure DI-H₂O and 5/95 PG/H₂O based nanofluids. The results were due to the following combinations: (a) the decreasing ζ values of 15/85 PG/H₂O nanofluids, which provide a less repulsive energy barrier; (b) the slightly higher A₁₃₁ value which increases the adhesive van der Waal force. Nevertheless, only the least stable nanofluid, 1 vol% 15/85 PG/H₂O, has W less than 10⁵ and is confirmed by the DLS results. Therefore, it is safe to say the tested nanofluids are stable during the experiments.

Table 3.3

Hamaker constantiii (A₁₃₁) and stability ratio of this study.

<table>
<thead>
<tr>
<th>Base fluid</th>
<th>A₁₃₁ [J]</th>
<th>Volume fraction</th>
<th>Stability ratio W</th>
</tr>
</thead>
<tbody>
<tr>
<td>DI-H₂O</td>
<td>4.33*10⁻²⁰</td>
<td>0.5%</td>
<td>1.78*10¹⁹</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.75%</td>
<td>7.59*10¹⁸</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1%</td>
<td>5.64*10¹⁸</td>
</tr>
<tr>
<td>5/95 PG/H₂O</td>
<td>5.19*10⁻²⁰</td>
<td>0.5%</td>
<td>2.61*10⁸</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.75%</td>
<td>7.21*10⁷</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1%</td>
<td>2.11*10⁷</td>
</tr>
<tr>
<td>15/85 PG/H₂O</td>
<td>5.41*10⁻²⁰</td>
<td>0.5%</td>
<td>2.65*10⁶</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.75%</td>
<td>4.83*10⁵</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1%</td>
<td>1.21*10⁴</td>
</tr>
</tbody>
</table>

iii Calculation of A₁₃₁ is attached in Appendix.
3.5. Summary

Nanoparticle and nanofluid characterizations including nanoparticle size in dry powder form and in suspensions, σ, pH and ζ-potential were presented in this chapter after a description of the nanofluid preparation processes. The TEM images show that dry nanoparticle powder has aggregations but the individual nanoparticles were spherical or nearly spherical by observation. The nanoparticle size in suspension was measured by a DLS system and the aggregation size shows an increasing trend with respect to time and nanofluid volume fraction. The stability ratio $W$ of nanofluid was also presented. Results showed that decreasing ζ-potential and increasing $A_{131}$ leaded the $W$ decreased with increasing nanoparticle loading and PG volume fraction.
Chapter 4: Pressure-drop viscosity measurement γ-\(\text{Al}_2\text{O}_3\) nanoparticles in water and propylene glycol-water mixtures

When particles are introduced in a liquid medium, the viscosity of the fluid is modified due to particle interactions or structure from the conditions of flow [30]. A colloidal solution with Newtonian behavior has usually been observed in liquids with dilute hard-sphere particles. For liquids with high polymer concentration, on the contrary, the rheological behavior of these solutions has often been reported as non-Newtonian [30]. The rheological behavior of a working fluid could affect the convective heat transfer outcome. This chapter is therefore focused on the nanofluid viscosity and first begins with a thorough literature review of nanofluid viscosity; it then presents results of base-fluid viscosity, followed by the comparison between measured and model evaluated nanofluid viscosity, and a short summary is also included in the end.

4.1. Literature review

The summary of published experimental nanofluid rehology studies is presented in Table 4.1. Prasher et al. [27] analyzed \(\text{Al}_2\text{O}_3\)-EG (ethylene glycol) nanofluids and observed that nanofluids behave as Newtonian fluids. By adopting the Krieger-Dougherty model, he showed that the size of aggregates was around 3 times the size of the individual nanoparticles. His analysis also pointed out, that in order to obtain a useful heat transfer enhancement of nanofluid relative to its base fluid, the viscosity
enhancement coefficient \((C_\mu)\) should be less than four times greater than the conductive enhancement coefficient \((C_k)\). i.e,

\[
C_\mu < 4C_k
\] (4.1)
<table>
<thead>
<tr>
<th>Ref.</th>
<th>Nanofluid</th>
<th>Particle size [nm]</th>
<th>Volume Fraction [%]</th>
<th>Shear Rate [s⁻¹]</th>
<th>Findings</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>[27]</td>
<td>Al₂O₃-PG</td>
<td>27, 40, 50</td>
<td>0.5, 2, 3</td>
<td>1~100</td>
<td>Newtonian behavior</td>
<td>Temperature: 30~60 °C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Viscosity increment is due to aggregates</td>
<td></td>
</tr>
<tr>
<td>[74]</td>
<td>CuO-H₂O</td>
<td>29</td>
<td>5, 8, 10, 13, 15</td>
<td>2~265</td>
<td>Pseudoplastic fluid.</td>
<td>Temperature: 5~50 °C</td>
</tr>
<tr>
<td>[75]</td>
<td>CuO-PG/H₂O</td>
<td>29</td>
<td>1, 2, 3, 4, 5, 5.9</td>
<td>50~275</td>
<td>Newtonian behavior</td>
<td>Temperature: -35~50 °C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Viscosity shows exponential function of ( \phi ) and temperature.</td>
<td></td>
</tr>
<tr>
<td>[76]</td>
<td>SiO₂-ethanol</td>
<td>35, 94, 190</td>
<td>1.1, 2.3, 3.1, 4.8, 5.1</td>
<td>1~50000</td>
<td>Newtonian behavior</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Good agreement with Krieger-Dougherty formula</td>
<td>35nm: 2.3, 5.1%</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>94nm: 3.1, 4.8%</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>190nm: 1.1%</td>
</tr>
<tr>
<td>[77]</td>
<td>CuO-EG/H₂O</td>
<td>29</td>
<td>1, 2, 3, 4, 5, 6.12</td>
<td>1.5~8</td>
<td>Newtonian behavior</td>
<td>Temperature: -35~50 °C</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Viscosity shows exponential function of ( \phi ) and temperature.</td>
<td></td>
</tr>
</tbody>
</table>
Table 4.1 (Cont’d)

Summary of experimental studies on nanofluid viscosity

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Nanofluid</th>
<th>Particle Size (nm)</th>
<th>Volume Fraction (%)</th>
<th>Shear Rate (s⁻¹)</th>
<th>Findings</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>[78]</td>
<td>TiO₂-EG</td>
<td>25</td>
<td>0~8 wt%</td>
<td>0.5~10⁴</td>
<td>Newtonian behavior Relative viscosity increment is independent of temperature</td>
<td>Temperature: 20~60 °C</td>
</tr>
<tr>
<td>[79]</td>
<td>TiO₂-EG</td>
<td>25</td>
<td>0.1, 0.21, 0.42, 0.86, 1.8</td>
<td>0.05~200</td>
<td>Newtonian behavior</td>
<td>Temperature: 20~60 °C</td>
</tr>
<tr>
<td>[80]</td>
<td>Al₂O₃-H₂O, CuO-H₂O</td>
<td>36, 47, 29</td>
<td>1~13</td>
<td>N/A</td>
<td>Viscosity depends on temperature and volume fraction. Particle size is important for Al₂O₃-H₂O for high ϕ. Hysteresis exists above critical temperature.</td>
<td>Temperature: 20~70 °C Dispersants were used⁴.</td>
</tr>
<tr>
<td>[81]</td>
<td>Al₂O₃-H₂O</td>
<td>30</td>
<td>0.02~0.3</td>
<td>N/A⁵</td>
<td>Viscosity increases nonlinearly with ϕ and the size of the nanoparticles relative to the tube size.</td>
<td>Temperature: 26 °C</td>
</tr>
</tbody>
</table>

⁴ Authors did not report chemical formulas of dispersants they used during experiments.
⁵ Laminar tube flow. Maximum Re is 75.
<table>
<thead>
<tr>
<th>Ref.</th>
<th>Nanofluid</th>
<th>Particle Size (nm)</th>
<th>Volume Fraction (%)</th>
<th>Shear Rate (s⁻¹)</th>
<th>Findings</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>[82]</td>
<td>Al₂O₃-C₁₀H₂₂</td>
<td>40</td>
<td>0.25~1</td>
<td>N/A</td>
<td>Shear viscosity exhibits an enhancement over the Einstein model.</td>
<td>Longitudinal viscosity was measured by impulsive stimulated thermal</td>
</tr>
<tr>
<td></td>
<td>Al₂O₃-PAO</td>
<td></td>
<td></td>
<td></td>
<td>Longitudinal viscosity data show nanoparticles do not aggregate in the</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>nanofluids.</td>
<td>scattering method.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>[83]</td>
<td>Fe₂O₃-H₂O</td>
<td>20~40</td>
<td>1~4</td>
<td>13.2~264</td>
<td>Nanofluids with φ&lt;0.02 behave as Newtonian fluids.</td>
<td>Used PEO or PVP as dispersants</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Temperature: 25 °C</td>
</tr>
</tbody>
</table>
References [74-83] experimentally show that the viscosity of nanofluids is strongly dependent on the nanoparticle volume fraction, temperature and size. Some particular features are pointed out as follows. Kulkarni et al. [74, 75] studied copper oxide (CuO) nanoparticles in H₂O and propylene glycol and water mixtures (PG/H₂O). For CuO in water [74], results showed that the nanofluids behave as shear thinning fluids for 5% < \( \phi < 15\% \). On the other hand, for CuO in PG/H₂O [75], the viscosity of nanofluids exhibited Newtonian behavior up to 5.9%. For Chen et al. [78, 79], the relative increase in the shear viscosity was only a function of \( \phi \) and was independent of temperature. The nanofluid viscosity of their lowest concentration sample (\( \phi = 0.5 \) wt\%) was less than that of its base fluid. The viscosity data for Al₂O₃-H₂O nanofluid of Nguyen et al. [80] appeared to exhibit hysteresis when the nanofluid sample was heated beyond some critical temperature. The authors suggested the hysteresis phenomenon might be due to particle agglomeration occurring at temperatures above this critical temperature, and hence leading to altered nanofluid properties. Another possible factor was the dispersants they used to prepare their nanofluids.

The shear and longitudinal viscosities of Al₂O₃ in C₁₀H₂₂ (decane) and isoparaffinic polyalphaolefin (PAO) were measured by Schmidt et al. [82]. The longitudinal viscosity was measured by an optical method, impulsive stimulated thermal scanning, together with thermal conductivity. The longitudinal viscosity depended on both volume fraction
and particle size, and therefore they were able to extrapolate the hydrodynamic diameter of the particles. Their results showed there were no clustered nanoparticles. Thus, the authors concluded that aggregation may not be responsible for the disagreement between the effective medium models and the measured nanofluid properties, which led these authors to conclude that the models based on Brownian motion or other nanoscale phenomena should be considered. The experimental data of Fe₂O₃-H₂O nanofluids with different dispersants were presented by Phuoc and Massoudi [83]. They found that for nanofluids having volume fractions greater than 2%, the viscosity has a shear thinning behavior.

Numerical studies of nanoparticle suspensions’ rheological behavior were conducted by In’t Veld [84] via a molecular dynamics method. They considered the effect of the solvent on the nanoparticle pair interaction by building up a solvent layer around the nanoparticles. The results showed that both nanoparticle size and particle solvent interaction are crucial if one wishes to treat the nanofluid as a continuum. Therefore, with increasing interaction strength, the effective viscosity also increased and may have shear thinning behavior. Masoumi [85] presented a model for calculating the effective viscosity of nanofluids by introducing the Brownian motion of nanoparticles into their model. Their model could calculate the effective viscosity as a function of temperature, particle diameter, particle density, volume fraction, and base fluid properties. The model has a good agreement with some other experimental data. However,
it should be noted that the model is based on the flow over a stationary spherical nanoparticle, which is $Re<<1$, the creeping flow assumption.

Based on this literature survey, it has been noticed that the viscosity of nanofluids in the published literature shows a spectrum of results, probably because several factors may affect nanofluids viscosity such as particle material, size, shape, carrier fluid, surfactant, flow velocity profile, etc. Therefore, the aim of this chapter is devoted to extending the scope of the measured viscosity of aqueous and propylene-glycol (PG) $\text{Al}_2\text{O}_3$-based nanofluids using our own experimental apparatus described in Chapter 2 and focus on the viscosity measurement of nanofluids in a ~ 1-mm tube under laminar flow conditions with or without constant heat flux applied at the tube surface.

4.2. Viscosity of base fluids

Before moving into nanofluid viscosity measurements, baseline conditions including viscosity of all base fluids under non-heating conditions are presented in Fig. 4.1. The data of pure $\text{H}_2\text{O}$ viscosities show very good agreement with tabulated values from reference [68]. The viscosities of 5/95 and 15/85 PG/$\text{H}_2\text{O}$ mixtures are also included in Fig. 4.1 and compared with data from measurements at Intel and from reference data from an ASHRAE handbook [67]. The PG/$\text{H}_2\text{O}$ mixtures measured by Intel were done by a Stresstech rheometer with a parallel plate fixture. It is observed that the discrepancy between our 5/95 PG/$\text{H}_2\text{O}$ measurement and ASHRAE is around 2.5%. For 15/85 PG/$\text{H}_2\text{O}$, our data
show a 16% difference from the reference data from the ASHRAE handbook. Compared with the data from Intel, the 5/95 and 15/85 PG/H₂O data show 9% and 3% differences, respectively. Note that since the ASHRAE handbook does not contain the exact same volume fraction as our test mixtures, we had to interpolate between two PG/H₂O volume fractions to obtain 5/95 and 15/85 PG/H₂O viscosities from [67].

![Fig. 4.1. Experimental values of base fluid viscosities.](image)

Table 4.2

<table>
<thead>
<tr>
<th>Volume Flow Rate [mL/min]</th>
<th>H₂O</th>
<th>5/95 PG/H₂O</th>
<th>15/85 PG/H₂O</th>
<th>H₂O</th>
<th>5/95 PG/H₂O</th>
<th>15/85 PG/H₂O</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>25°C</td>
<td></td>
<td></td>
<td>Apply constant heat flux at wall</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>0.8845</td>
<td>0.9226</td>
<td>1.7579</td>
<td>0.6609</td>
<td>0.6356</td>
<td>1.0458</td>
</tr>
<tr>
<td>5</td>
<td>0.8933</td>
<td>0.9199</td>
<td>1.7816</td>
<td>0.7298</td>
<td>0.6851</td>
<td>1.1289</td>
</tr>
<tr>
<td>9</td>
<td>0.8907</td>
<td>0.9239</td>
<td>1.7802</td>
<td>0.7688</td>
<td>0.7450</td>
<td>1.2116</td>
</tr>
</tbody>
</table>
The base fluid viscosities of all cases are presented in Table 4.2. Note that since the wall boundary has a constant heat flux condition, the test fluid at a lower volume flow rate has a higher temperature and thus lower viscosity, given the general trend that viscosity decreases with increasing temperature.

### 4.3. Results and discussion of nanofluid viscosity data

The viscosity hereafter will be reported as relative viscosity ($\mu_r$) defined as

\[
\mu_r = \frac{\mu_{nf}}{\mu_b}
\]  

(4.2)

where $\mu_{nf}$ is the effective viscosity of nanofluids measured under specific conditions and $\mu_b$ is the effective viscosity of the base fluid under the same conditions, i.e., same volume flow rate, heating condition, and temperature.

Effects of the volume flow rates and particle volume fractions on nanofluids $\mu_r$ are shown as Figs. 4.2 - 4.4. Fig. 4.2 demonstrates the viscosity values of 20-nm Al$_2$O$_3$ nanoparticles in DI- H$_2$O. The viscosity data of 20-nm Al$_2$O$_3$ nanoparticles in 5/95 and 15/85 PG/H$_2$O are presented in Figs. 4.3 and 4.4.
Fig. 4.2. Experimental results of relative viscosity of 20-nm Al$_2$O$_3$-H$_2$O nanofluids with different volume fraction. The ‘H’ curves represent measurements under heated wall conditions.

Fig. 4.3. Experimental results of relative viscosity of 20-nm Al$_2$O$_3$ nanoparticles in 5/95 PG/H$_2$O with different volume fraction.
Fig. 4.4. Experimental results of relative viscosity of 20-nm Al₂O₃ nanoparticles in 15/85 PG/H₂O with different volume fraction.

The relative viscosities for each nanofluid volume fraction for the unheated cases (Figs. 4.2 – 4.4) do not show any apparent trends with respect to volume flow rate, but fluctuates about a mean value. There is a distinct trend that the $\mu_r$ of nanofluids increases with increasing particle volume concentration and the trend can be clearly seen in Fig. 4.5 which plots the $\mu_r$ of nanofluid against the nanofluid volume fraction. The trend that $\mu_r$ increases with increasing volume fraction is expected.
Fig. 4.5. Experimental values for 20-nm Al$_2$O$_3$ nanoparticles in different base fluids for a volume flow rate of 5 mL/min. The ‘H’ curves represent measurements under heated wall conditions.

For laminar flow in a circular tube, the minimum shear rate is zero at the centerline and the maximum shear rate occurs at the wall ($\tau_w$) which can be described as

$$\tau_w = \frac{D \Delta P}{4L}$$  \hspace{1cm} (4.3)

Among all cases, the wall shear stress fell within the range of 1 to 35 s$^{-1}$. Furthermore, once particles are suspended in a liquid, it is likely that they organize into a structure which makes the liquid layer around the particle surface less mobilized [30]. This retardation makes the liquid more viscous and when a shear stress is imposed the liquid usually exhibits shear-thinning behavior as a consequence, in which the viscosity decreases
with increasing shear stress. For non-heated cases, there is no such shear-thinning appearance for all tested nanofluids. Plus all tested nanofluids have $\phi$ less than or equal to 1 vol%. Therefore, it is possible that the nanofluids in all our experiments did not go through a structural deformation because of shear forces, and it is safe to assume the nanofluids exhibited Newtonian behavior during all experiments.

For cases with applied constant wall heat flux, the data is denoted by a red ‘H’ curve in all graphs. It is shown that the relative viscosity decreases with increasing volume flow rate when constant wall heat flux was applied. The relative viscosities with the heater on were greater than the cases without constant heat flux, especially for the 1 mL/min and 5 mL/min experiments. This phenomenon could be the result of the temperature increase. First, when temperature increases above room temperature, the nanoparticles acquire more energy and have a higher Brownian speed. Nanoparticles under such conditions appear to have rather high particle interactions and increased collision frequency. The nanofluid viscosity increases with increasing particle interactions [86]. The higher collision rate can also increase the nanoparticles’ aggregation probability and hence the aggregates can give rise to additional flow resistance. Second, with the constant heat flux on the test tube, the nanofluids temperature keeps growing after entering the test section. As a given packet of nanofluids proceeds downstream, the nanoparticles are exposed to higher temperatures and have higher energy compared with
the nanoparticles at an upstream location. So the nanoparticles tend to move towards the cold upstream end due to thermophoresis, which also increases the resistance to flow. The viscosity is therefore increased.

In Fig. 4.6, the measured $\mu_r$ of 20-nm Al$_2$O$_3$-H$_2$O nanofluids were compared with Einstein’s model [31] and the correlation by Prasher et al. [27]. The analysis of [27] starts with the Krieger-Dougherty model [87], where the aggregates size and $\phi$ are inserted into the Krieger-Dougherty expression. Their analysis shows that the ratio of the aggregates’ radius to that of a single nanoparticle is 3.17 and the relative viscosity of nanofluids becomes

$$\mu_r = 1 + C_\mu \phi = 1 + 10\phi$$

(4.4)

This equation shows that when the size of the aggregates is around three times the size of an individual nanoparticle, the $C_\mu$ coefficient will be about four times the value from Einstein’s model, eq. (1.1). The correlation of Prasher et al. showed good agreement with published nanofluid viscosity data [74, 79]. So, the correlation from [27] is chosen to compare with this study.
Fig. 4.6. Comparison of 20-nm Al₂O₃-H₂O experimental values with Einstein’s model [31] and the Prasher et al. experimental correlation [27]. Relative viscosities of long thin prolate spheroids predicted by Brenner [86] are also included. The ‘H’ data represent measurements under heated wall conditions, and ‘aₚ’ is the particle axis ratio.

As shown in Fig. 4.6, the experimental data are much greater than Einstein’s prediction for both unheated and heated cases. There is much better agreement with the Prasher et al. expression [27], but generally our data lie higher than that correlation, especially for higher volume fractions. Both the Einstein model [31] and the correlation from Prasher et al. [27] do not include the effect of temperature, so both the heated and unheated cases share the same prediction.
Additional information regarding the nanoparticle aggregate shape effect is also provided in Fig. 4.6. For nanofluids flowing in a circular tube within the laminar flow regime, it is possible that particles coagulate into chains or prolate spheroid-shape clumps due to the shear force acting on the nanoparticles and clusters. For uniformly suspended particles, the suspensions with long and slender-shaped aggregates have higher viscosity and exhibit more complex flow behaviors compared with those with spherical aggregates [86]. Additionally, if chains or spheroid-shape aggregates occur in the flow field, the aggregates can not rotate freely in the flow field when shear stress is applied. Therefore, the lack of degree of freedom of the aggregates will dramatically increase the suspension viscosity. In order to calculate the aggregation size, the model from Brenner’s work [86] was adopted and the aggregation shape was assumed to be long thin prolate spheroids. In [86], the rheology of suspensions can be modeled by nondimensional scalar material constants which contain information on the size, shape and $\phi$ of suspended particles or aggregates. The viscosity of long thin spheroids suspended in liquids can be described as [86]

$$\mu_r = 1 + \phi(5Q_1 - Q_2 + 2Q_3)$$

(4.5)

where

$$Q_1 = \frac{2}{5} - \frac{6\ln 2a_p}{5a_p^2}$$

(4.6)
\[ Q_2 = \frac{2}{5} - \frac{a_p^2}{12(\ln 2a_p - 1.5)} \]  

(4.7)

\[ Q_3 = \frac{a_p^2}{10(\ln 2a_p - 0.5)} \]  

(4.8)

and \( a_p \) is the particle axis ratio given as

\[ a_p = \frac{a}{b} \]  

(4.9)

where \( a \) is the length of the polar radius and \( b \) is the equatorial radius.

Based on Brenner’s model, it appears that the heated cases of 20-nm Al₂O₃ nanoparticles in all tested base fluids have relatively elongated aggregates due to the higher \( \mu_r \) we observed, especially at lower flow rates (Figs. 4.2-4.4). Again, the temperature could play an important role in this case. When the base fluid temperature increases, the viscosity of the base fluid decreases. The surroundings now provide the nanoparticles a higher probability to overcome inter-particle repulsion and coagulate into chains when under shear. When comparing different nanofluid volume fractions at the same fluid flow rate like in Fig. 4.6, the higher nanoparticle volume fractions always results in a higher relative viscosity. From Fig. 4.6, the limited data suggests that \( a_p \) increases slightly with increasing \( \varphi \) for unheated nanofluids; while for the heated nanofluids \( a_p \) remains constant or slightly decreases with increasing \( \varphi \).
4.4. Summary

The viscosity of 20-nm, γ-Al2O3 water-based and water/propylene glycol-based nanofluids in a single 1.02-mm inner diameter, on/off constant heat flux stainless steel tube, was experimentally investigated for laminar flow. Overall, experimental results show that the relative viscosity, $\mu_r$, increases with nanoparticle volume fraction. For unheated cases, our data did not show any conclusive trend with respect to nanofluid volume flow rate. There may be some difference in how the aggregate aspect ratio changes with increasing volume fraction, depending on whether or not the tube is heated. For the cases with applied constant heat flux, $\mu_r$ showed decreasing trends with respect to increasing volume flow rate. In general, nanofluids flowing in heated tubes exhibited higher $\mu_r$, which may be due to highly frequent particle interactions as the nanofluid temperature increases, and possibly to the effect of thermophoretic flow from the hotter downstream end to the cooler upstream end.
Chapter 5: Convective heat transfer coefficient of alumina nanofluids

The up-to-date experimental nanofluid convective heat transfer studies under laminar flow condition is presented in Table 5.1. It has been noticed that most groups focused on the laminar flow region of 500<Re<2000. All researches returned positive results on heat transfer coefficient when using nanofluids as working fluids, i.e. $h_{\text{nanofluid}}>h_{\text{base fluid}}$, regardless of the different combinations of nanoparticle material, nanoparticle shape, base fluid, boundary conditions and so on. Only a few groups, however, reported their results in nondimensional form, i.e., as a Nusselt number $Nu = hD/k_{nf}$, where $D$ is a length scale, and $k_{nf}$ is the nanofluid thermal conductivity [34, 35, 38, 46, 47 and 88]. In these researches, most authors report favorable $Nu$ results which indicate that $h$ increases relative to the increase in $k_{nf}$. However, when comparing results from different groups, the magnitude of the augmentation in $h$ and $Nu$ does not show any agreement so far. Additionally, several contradictory results were observed. Yang et al. [35] observed that the ratio of nanofluid to base fluid $h$ is less than the ratio of $k$ between nanofluid and its base fluid. Therefore, the results in [35] should have nanofluid’s $Nu$ less than its base fluid. Jung et. al. [46] studied forced convective heat transfer of nanofluids in microchannels for $Re$ within 5 to 300 and their results showed that all $Nu$ results, including base fluids, are less than 0.5. Their results are substantially less than theoretical findings in microchannels.
[90] and imply that nanofluids may not be applicable in microchannels. Rea et. al. [47] tested Al₂O₃ and ZrO₂ nanofluid in thermally developing region. Their Nu results exhibited good agreement with traditional predictions and hence they concluded nanofluids behave as homogeneous mixtures.

This chapter presents all the experimental data regarding convective heat transfer coefficient data of nanofluids, taken during the course of this dissertation research, in dimensional form which is in units of Wm⁻²K⁻¹. First is the experimental result of DI-H₂O based nanofluids from the test rig introduced in chapter 2. The second part is the convective heat transfer data of 5/95 PG/H₂O based nanofluids. The data of 15/85 PG/H₂O based nanofluids are presented in the third section and a discussion of possible enhancement mechanisms is presented in section 5.4. And finally, a short summary is provided in section 5.5.
Table 5.1
Summary of experimental studies on convective heat transfer performance of nanofluid under laminar flow region

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Nanofluids (Surfactant)</th>
<th>Particle Size (nm)/ Shape</th>
<th>Concentration</th>
<th>Flow Regime ($Re$)</th>
<th>Findings</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>[34]</td>
<td>γ-Al$_2$O$_3$ + H$_2$O (Sodium dodecylbenzene sulfonate)</td>
<td>27-56 / spherical</td>
<td>0.6~1.6 vol%</td>
<td>500~2000</td>
<td>$h &gt; h_{H2O}$ $Nu &gt; Nu_{H2O}$</td>
<td>Developing region. Constant wall temperature.</td>
</tr>
<tr>
<td>[35]</td>
<td>Graphite+ commercial automatic transmission fluid or mixture of two commercial oils with additives.</td>
<td>d:1-2µm &amp; δ:20-40nm/ disk-like</td>
<td>2 &amp; 2.5 wt%</td>
<td>5~80</td>
<td>$h &gt; h_{base fluid}$ $h/h_{base fluid} &lt; k_{nf}/k_{base fluid}$</td>
<td>Constant wall temperature.</td>
</tr>
<tr>
<td>[36]</td>
<td>CNT + H$_2$O (Gum Arabic)</td>
<td>&gt;100nm/ rod-like</td>
<td>0.1 ~ 0.5 wt%</td>
<td>800~1200</td>
<td>$h &gt; h_{base fluid}$</td>
<td>Developing region</td>
</tr>
<tr>
<td>[37]</td>
<td>Cu + H$_2$O</td>
<td>50-60 nm/ N/A</td>
<td>0.2~3.0 vol%</td>
<td>650~2050</td>
<td>$h &gt; h_{H2O}$</td>
<td>Constant wall temperature.</td>
</tr>
<tr>
<td>[39]</td>
<td>γ-Al$_2$O$_3$ + H$_2$O</td>
<td>20 nm/ spherical</td>
<td>0.2~2.5 vol%</td>
<td>700~2050</td>
<td>$h &gt; h_{H2O}$ $Nu &gt; Nu_{H2O}$</td>
<td>Constant wall temperature.</td>
</tr>
</tbody>
</table>
Table 5.1. (Cont’d)

Summary of experimental studies on convective heat transfer performance of nanofluid under laminar flow region

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Nanofluids (Surfactant)</th>
<th>Particle Size (nm)/Shape</th>
<th>Concentration</th>
<th>Flow Regime ((Re))</th>
<th>Findings</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>[41]</td>
<td>CuO + H(_2)O</td>
<td>d:80 &amp; l:20nm/needle</td>
<td>0.204~0.4 vol%</td>
<td>(V =10, 15 &amp; 20) ml/min(^6)</td>
<td>(h&gt;h_{H_2O})</td>
<td>performance(<em>{nf}&gt;)performance(</em>{H_2O}) Microchannel</td>
</tr>
<tr>
<td>[42]</td>
<td>Titanate+ H(_2)O</td>
<td>d:10 &amp; l:100nm/rod-like</td>
<td>0.5~2.5 wt%</td>
<td>1100~2300</td>
<td>(h&gt;h_{H_2O})</td>
<td>Constant wall heat flux.</td>
</tr>
<tr>
<td>[45]</td>
<td>Al(_2)O(_3) + H(_2)O</td>
<td>30±5 nm/spherical</td>
<td>0.01~0.3%</td>
<td>550~750</td>
<td>(h&gt;h_{H_2O})</td>
<td>(h/h_{H_2O}) is about constant Fully developed region. Constant wall heat flux.</td>
</tr>
<tr>
<td>[46]</td>
<td>Al(_2)O(_3) + H(_2)O</td>
<td>170±10 nm/ N/A</td>
<td>0.6~1.8 vol%</td>
<td>5~300</td>
<td>(h&gt;h_{\text{base fluid}}) Nu increases with increasing (Re) Nu&lt;0.5</td>
<td>Microchannel</td>
</tr>
</tbody>
</table>

\(^6\) \(Re\) numbers were not provided.
Table 5.1. (Cont’d)

Summary of experimental studies on convective heat transfer performance of nanofluid under laminar flow region

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Nanofluids (Surfactant)</th>
<th>Particle Size (nm)/Shape</th>
<th>Concentration</th>
<th>Flow Regime ((Re))</th>
<th>Findings</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>[47]</td>
<td>(\text{Al}_2\text{O}_3 + \text{H}_2\text{O}) (\text{ZrO}_2 + \text{H}_2\text{O})</td>
<td>50nm/N/A</td>
<td>0.6<del>6 vol% 0.32</del>1.32 vol%</td>
<td>100~2000</td>
<td>(h &gt; h_{\text{H}_2\text{O}}) (Nu) show good agreement with theoretical prediction</td>
<td>Constant wall heat flux Developing region</td>
</tr>
<tr>
<td>[49]</td>
<td>(\text{Al}_2\text{O}_3 + \text{H}_2\text{O})</td>
<td>45 &amp; 150nm/Spherical</td>
<td>1~6 wt%</td>
<td>700~2100</td>
<td>(h &gt; h_{\text{H}<em>2\text{O}}) (Nu &gt; Nu</em>{\text{H}_2\text{O}}) Nanofluid with 45nm nanoparticles has higher (h) than Nanofluid with 150nm nanoparticles.</td>
<td>Constant wall heat flux Developing region Control stability of nanofluids by (pH).</td>
</tr>
<tr>
<td>[53]</td>
<td>Amorphous carbonic+ (\text{H}_2\text{O}) (\text{Al}_2\text{O}_3 + \text{H}_2\text{O})</td>
<td>20nm/Spherical 20-50nm/Spherical</td>
<td>3.5 vol% 3 vol%</td>
<td>800~6500</td>
<td>(h &gt; h_{\text{H}_2\text{O}})</td>
<td>Constant wall heat flux</td>
</tr>
</tbody>
</table>
Table 5.1. (Cont’d)

Summary of experimental studies on convective heat transfer performance of nanofluid under laminar flow region

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Nanofluids (Surfactant)</th>
<th>Particle Size (nm)/ Shape</th>
<th>Concentration</th>
<th>Flow Regime ($Re$)</th>
<th>Findings</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>[54]</td>
<td>Diamond+ H$_2$O Diamond+ EG</td>
<td>30~50nm/Spherical</td>
<td>0.5~2 vol%</td>
<td>Re&lt;1800</td>
<td>$h &gt; h_{base fluid}$ 2 vol% nanofluid has $h$ between base fluid and other nanofluids</td>
<td>Constant wall temperature.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>[55]</td>
<td>MWCNT + H$_2$O (Gum Arabic)</td>
<td>d: 10<del>20nm &amp; l: 0.5</del>40 μm/ Rod-like</td>
<td>0.25 wt%</td>
<td>600~1200</td>
<td>$h &gt; h_{base fluid}$ Optima sonication time is 40 minutes.</td>
<td>Constant wall heat flux</td>
</tr>
</tbody>
</table>


Table 5.1. (Cont’d)

Summary of experimental studies on convective heat transfer performance of nanofluid under laminar flow region

<table>
<thead>
<tr>
<th>Ref.</th>
<th>Nanofluids (Surfactant)</th>
<th>Particle Size (nm)/ Shape</th>
<th>Concentration</th>
<th>Flow Regime (Re)</th>
<th>Findings</th>
<th>Note</th>
</tr>
</thead>
</table>
| [88] | Al$_2$O$_3$ + transformer oil (Oleic acid)  
Al$_2$O$_3$+transformer oil (Oleic acid)  
AlN + transformer oil (Oleic acid)  
(Ho Wong)  
Kwok et al.  
2004  
100–500  
h$>$h$_{base$ fluid | 13nm/Spherical  
d: 2nm & l: 20–200nm/Rod-spherical  
50nm/Spherical | 0.5 vol% | 100–500 | h$>$h$_{base$ fluid | In plate type heat exchanger |
| [89] | TiO$_2$+ H$_2$O (Cetyl trimethyl ammonium bromide)  
(Kwok et al.  
2004  
100–500  
h$>$h$_{H_2O}$  
Nu$>$ Nu$_{H_2O}$ | 15nm/Spherical | 0.2–0.8 vol% | 900–1700 | h$>$h$_{H_2O}$  
Nu$>$ Nu$_{H_2O}$ | Constant wall heat flux. Convective heat transfer result of nanofluid only compare with pure DI-H$_2$O data. |
5.1. Convective heat transfer results of DI-H₂O based nanofluid

The local heat transfer coefficients in the thermally developing region are presented as a function of axial distance from the test tube entrance, at three different volume flow rates, in Fig. 5.1 (a), (b) and (c), respectively. Most $h$ results for the nanofluids are greater than that of pure DI-H₂O except for the nanofluid with volume fraction 0.5% at the entrance. In Fig. 5.1, several trends can be observed. First of all, $h$ decreases with increasing axial distance from the test tube entrance. Secondly, the $h$ results of the Al₂O₃-DI-H₂O nanofluid show that $h$ increases with an increasing flow rate and particle volume fraction. Additionally, the enhancements in $h$ near the entrance are greater than the enhancements in $h$ further downstream. Finally, the nanofluids exhibit a longer entrance region than pure DI-H₂O.
Fig. 5.1. (a) Local heat transfer coefficients of DI-H₂O based nanofluids in the thermal developing region at 1 mL/min volume flow rate.

Fig. 5.1 (b) Local heat transfer coefficients of DI-H₂O based nanofluids in the thermal developing region at 5 mL/min volume flow rate.
Fig. 5.1. (c) Local heat transfer coefficients of DI-H\textsubscript{2}O based nanofluids in the thermal developing region at 9 mL/min volume flow rate.

Fig. 5.2. Heat transfer coefficient of water-based 20-nm Al\textsubscript{2}O\textsubscript{3} nanofluids in the fully developed region.
Figure 5.2 presents $h$ for nanofluids under different flow rates in the fully developed region. In the fully developed region, the theoretical prediction [61] for pure DI- H$_2$O gives the theoretical $h$ values by using eq. (2.10) along with $Nu(x) = 4.364$ for constant heat flux at the tube surface. These calculated $h$ values for pure DI- H$_2$O show a gradually decreasing trend, which is due to the temperature dependence of $k_{H_2O}$ as mentioned before. The $h$ results for the nanofluids did not show the same trend. Instead, $h$ for the nanofluids tends to increase with increasing volume flow rate and particle volume fraction. However, at low volume flow rates, the $h$ results for the 0.5% and 0.75% nanofluids are nearly the same. Moreover, the enhancements in $h$ increased with increasing particle volume fraction.

5.2. Convective heat transfer results of 5/95 PG/H$_2$O based nanofluid

The local heat transfer coefficients in the thermally developing region are presented as a function of axial distance from the test tube entrance, at three different volume flow rates, in Fig. 5.3 (a), (b) and (c), respectively. The $h$ of 5/95 PG/H$_2$O based nanofluids share a similar trend with DI-H$_2$O based nanofluids in the developing region. All 5/95 PG/H$_2$O based nanofluids $h$ data are less than those of DI-H$_2$O based nanofluids. The results are expected because, compared with DI-H$_2$O based nanofluids, 5/95 PG/H$_2$O based nanofluids are more viscous and have less thermal conductivity. Figure 5.4 presents $h$ for 5/95 PG/H$_2$O based nanofluids under different flow rates in the fully developed region.
In the fully developed region, the 5/95 PG/H₂O based nanofluids $h$ seems mostly depending on nanoparticle loading, otherwise $h$ decreasing with increasing volume flow rate.

Fig. 5.3. (a) Local heat transfer coefficients of 5/95 PG/H₂O based nanofluids in the thermal developing region at 1 mL/min volume flow rate.
Fig. 5.3. (b) Local heat transfer coefficients of 5/95 PG/H$_2$O based nanofluids in the thermal developing region at 5 mL/min volume flow rate.

Fig. 5.3. (c) Local heat transfer coefficients of 5/95 PG/H$_2$O based nanofluids in the thermal developing region at 9 mL/min volume flow rate.
Fig. 5.4. Heat transfer coefficient of 5/95 PG/H₂O based 20-nm Al₂O₃ nanofluids in the fully developed region.
5.3. Convective heat transfer results of 15/85 PG/H$_2$O based nanofluid

The local heat transfer coefficients in the thermally developing region are presented as a function of axial distance from the test tube entrance, at three different volume flow rates, in Fig. 5.5 (a), (b) and (c), respectively. Figure 5.6 presents $h$ for nanofluids under different flow rates in the fully developed region. The $h$ results of 15/85 PG/H$_2$O based nanofluids only show distinguishable enhancement at the entrance then $h$ quickly drops along the flow direction and the enhancement becomes insignificant. It also shows that compared with the previous two sets of data, 15/85 PG/H$_2$O based nanofluids have shorter thermal entrance length. In the fully developed region, it seems nanofluids have very little enhancement on $h$ and only 1 vol% 15/85 PG/H$_2$O shows increasing trend with regard to increasing volume flow rate.
Fig. 5.5. (a) Local heat transfer coefficients of 15/85 PG/H₂O based nanofluids in the thermal developing region at 1 mL/min volume flow rate.

Fig. 5.5. (b) Local heat transfer coefficients of 15/85 PG/H₂O based nanofluids in the thermal developing region at 5 mL/min volume flow rate.
Fig. 5.5. (c) Local heat transfer coefficients of 15/85 PG/H₂O based nanofluids in the thermal developing region at 9 mL/min volume flow rate.

Fig. 5.6. Heat transfer coefficient of 15/85 PG/H₂O based 20-nm Al₂O₃ nanofluids in the fully developed region.
5.4. Discussion

The overall mechanisms by which the presence of nanoparticles can enhance the static thermal conductivity, relative to its base fluid, have been studied by many research groups, but there remains significant debate over both the mechanisms and the extent of enhancement [18]. One possible enhancement mechanism could be that the observed increases in $h$ are due entirely to increases in the static thermal conductivity of nanofluids, $k$. Another possibility, suggested a number of years ago by Ahuja [4, 5] for micron-size particles, is that particle rotation could also lead to convective heat transfer augmentation. That is to say, when particles under the effect of shear stress created by the surrounding flow filed, particles perpendicularly rotate about the axis to the main flow direction. Hence, the greater the velocity gradient along the radiant direction of the test tube, the greater the particle rotating velocity and the resulted enhancement will be. Wen and Ding [34] proposed that the enhancement might be due to particle migration within the flow field, especially near the entrance where a larger velocity gradient is expected. Particle size and shape are also parameters which might affect the heat transfer enhancement because Yang et al. [35] observed only a very small enhancement when using disk-like particles.

The observed greater enhancement at the entrance region could be due to how the thermal boundary layer develops for a nanofluid compared to a pure fluid. From [57] and [61], $h$ decreases with increasing thermal
boundary thickness and remains constant after approaching the fully developed condition. $h$ can be approximated by $h \sim k_{nf}/\delta$, where $k_{nf}$ is the nanofluid thermal conductivity and $\delta$ the thermal boundary layer thickness. Therefore, either increasing $k_{nf}$ or reducing $\delta$ can increase $h$.

Researches by Prasher et al. [27] showed that the Prandtl number ($Pr$), $Pr = c\mu k^{-1}$, for nanofluids increases with increasing volume fraction. Per the classic entrance-region length given in eq. (2.11), a greater $Pr$ means a longer entrance region, or thus a smaller $\delta$ at a given location $x$ compared to that of the pure liquid. This trend is borne out by the 1 vol% Al$_2$O$_3$-DI-H$_2$O nanofluid curves in Fig. 5.2, which appear to exhibit longer entrance regions than the other curves. Hence, greater enhancement in $h$ in the entrance region for nanofluids can be explained, at least in part, by classic $Pr$-number effects.

The mechanisms responsible for the observed enhancement in $h$ in the fully developed region might be not only due to changes in the effective thermophysical properties by the presence of nanoparticles (i.e., thermal conductivity and viscosity), but also due to nanoparticle dispersion. The idea was proposed by Buongiorno [91] using a scaling analysis. However, the analysis in [91] emphasizes the importance of the local nanoparticle distribution within the fluid by including a “nanoparticle continuity” equation. Mills and Snabre [92] studied the particle concentration distribution in the case of Poiseuille flow in a circular pipe. Their results showed that suspensions exhibit a higher concentration core at the center.
of the channel and the concentration distribution depends on the suspension volume fraction. With a higher concentration core, the nanofluid viscosity around the centerline of the circular tube is greater than the viscosity away from the centerline; the velocity profile is flattened around the centerline, but has a steeper gradient near the wall. Therefore, $h$ increases. Regarding the thermal conductivity distribution along the radial direction, Sohn and Chen [93] correlated the slurry thermal conductivity as a function of shear rate, which is proportional to radial distance, and showed significant enhancement in $h$ even though they assumed a parabolic velocity profile in the first place.

5.5. Summary.

The convection heat transfer performance of 20-nm, $\gamma$-Al$_2$O$_3$ nanoparticles in three different base fluids in a single 1.02-mm inner diameter, constant heat flux stainless steel tube, was experimentally investigated for laminar flow, in both the developing and fully developed regions. It is evident that nanofluids give higher $h$ than the base fluids. Overall, experimental results show that the heat transfer coefficient, $h$, increases with volume flow rate and nanoparticle volume fraction. The $h$ enhancements also increased with increasing volume flow rate and nanoparticle volume fraction. The $h$ enhancement is significant in the entrance region and decreased with increasing axial distance from the test section entrance. These results also showed that the higher the volume fraction, the longer is the thermal entrance length.
Chapter 6: Nanofluid convective heat transfer performance in nondimensional form

It is of interest to compare the convective and conductive performances of nanofluids under laminar flow conditions in nondimensional form, via the classical Nusselt number \((Nu)\). The \(Nu\) results of nanofluids will be plotted against Reynolds number \( (Re)\), defined as

\[
Nu(x) = \frac{h(x)D_l}{k} \quad (6.1)
\]

\[
Re = \frac{\rho U_m D_l}{\mu} \quad (6.2)
\]

where \(k\) is the effective nanofluid thermal conductivity and \(\mu\) is the effective nanofluid viscosity which is obtained from Chapter 4.

In order to calculate \(Nu\), the model and results for nanofluid \(k\) are presented in section 6.1. The \(Nu\) of all nanofluids in the thermally developing region will be presented in section 6.2. The third part will be the \(Nu\) of all nanofluids in the fully developed region. The discussion and a summary will be presented in the end. Note that all convective heat transfer data were measured under laminar flow conditions. Also note that all nanofluid thermal properties were determined at the average temperature of the nanofluid, which is justified here over the limited temperature range of our experiments (20-45 °C).
6.1. Calculation of nanofluid thermal conductivity

6.1.1 Thermal conductivity model

Since nanoparticles tend to aggregate into clusters [29] a consensus has emerged in the literature that the increase in $k$ is due to percolation effects within the aggregates, when highly conducting particles touch each other and create high conductivity paths. Therefore, the following computation of nanofluid $k$ is based on the model from Prasher et al. [62] who consider aggregation-kinetics effects in their thermal conductivity model. The general idea is to first calculate the effective thermal conductivity of a cluster ($k_a$) and then calculate the overall thermal conductivity ($k$) of nanofluid by a Maxwell-Garnett type model (M-G model). The $k_a$ can be solved via the Bruggeman model [94]:

$$\phi_{int}\left(\frac{k_p-k_a}{k_p+2k_a}\right)+(1-\phi_{int})\left(\frac{k_b-k_a}{k_b+2k_a}\right)=0$$

(6.3)

where $k_p$ is the nanoparticle thermal conductivity and $k_b$ is the base fluid thermal conductivity. Furthermore, $\phi_{int}$ is the ratio of the volume of nanoparticles to the volume of aggregate. Once $k_a$ is determined, the $k$ of nanofluid is [95]

$$\frac{k}{k_b} = \frac{k_a + 2k_b + 2\phi_a(k_a-k_b)}{k_a + 2k_b - \phi_a(k_a-k_b)}$$

(6.4)

where $\phi_a$ is the volume fraction of aggregates in the entire suspension. If the nanoparticles are completely dispersed in the suspension, eq. (6.4) will reduce to the M-G model which will then neglect any interfacial thermal
resistance between nanoparticles and base fluid. One can relate $\phi_a$ and $\phi_{int}$ via the following equation:

$$\phi_p = \phi_{int}\phi_a$$  \hspace{1cm} (6.5)

Assume at the beginning (time, $t=0$) that the nanoparticles are individually well dispersed and have a volume fraction of $\phi_p$. Therefore, from eq. (6.5) $\phi_{int}=1$ represents that there is only one particle in each aggregate and $\phi_a=\phi_p$. Eventually, all particles may form one big aggregate, then $\phi_a=1$ and $\phi_{int}=\phi_p$. At a certain intermediate time ($t$), nanoparticles agglomerate into multiple well-dispersed clusters as shown in Fig. 6.1 where the gyration radius, $R_a$, can be viewed as the effective radius of an aggregate and the relation between $R_a$ and $\phi_{int}$ is

$$\phi_{int} = \left(\frac{R_a}{r_p}\right)^{d_f-3}$$  \hspace{1cm} (6.6)

where $d_f$ is the fractal dimension. Generally, $d_f$ ranges from 2 to 3 for dense aggregates and falls below 2 for open ones. Wang et al. [96] observed $d_f=1.8$ for diffusion-limited cluster-cluster aggregation (DLCCA) of nanofluids. Therefore, $d_f$ will be set to 1.8 here, as was done in [62]. $r_p$ is the radius of the individual nanoparticles and equals to 20 nm from the manufacturer's specification. Because nanoparticle aggregation is time dependent, the $R_a$ needs to be correlated with the aggregation time constant ($t_p$) which characterizes the time for cluster formation and can be related to nanofluid stability ratio ($W$), given previously in Table 3.3:
Fig. 6.1. Schematic diagram of well dispersed clusters. $R_a$ is the gyration radius of aggregates and the red curve indicates high thermal conductivity-percolation path.

\[
\frac{R_a}{r_p} = \left(1 + \frac{t}{t_p}\right)^{\frac{1}{d_f}}
\]

(6.7)

Substituting eq. (6.7) into eq. (6.6) yields

\[
\phi_{int} = \left(1 + \frac{t}{t_p}\right)^{\frac{d_f - 3}{d_f}}
\]

(6.8)

and $t_p$ can be written as [72]

\[
t_p = \frac{\pi r_p^3 \mu W}{\phi_p k_B T}
\]

(6.9)

where $k_B$ is the Boltzmann constant.

6.1.2. Results for nanofluid thermal conductivity

The computed $t_p$ results for nanofluids are presented in Table 6.1. As the nanoparticle concentration or PG concentration increases, the $t_p$ results have the same decreasing trend as $W$. It is also shown that, with a small decrease in $W$, $t_p$ could dramatically decrease especially for those
nanofluids based on PG/H\textsubscript{2}O. This phenomenon is expected for those nanofluids with higher value of Hamaker constant, \(A_{131}\), and lower zeta potential, \(\zeta\). This combination implies greater attractive force and less repulsive force. Therefore, the values for \(t_p\) significantly decrease even though the 1 vol\% Al\textsubscript{2}O\textsubscript{3} 15/85 PG/H\textsubscript{2}O nanofluid has \(W>>1\) and was assumed stable in the first place.

The predicted thermal conductivity ratio versus time is presented in Figs. 6.2-6.4. The computed \(k\) needs to coincide with the period of time when the convection experiments were conducted. In this study, \(t=0\) is assumed to be the time the nanofluids come right out of the ultrasonicator. Convection experiments were carried out after 1 hour and the experiment duration is approximately 1 hour for each nanofluid. Hence the applicable value of \(k\) is marked by a band from 1 to 2 hours after \(t = 0\).

Table 6.1

<table>
<thead>
<tr>
<th>Base fluid</th>
<th>Volume fraction</th>
<th>Aggregation time constant (t_p) [hr]</th>
</tr>
</thead>
<tbody>
<tr>
<td>DI-H\textsubscript{2}O</td>
<td>0.5%</td>
<td>5.38*10\textsuperscript{12}</td>
</tr>
<tr>
<td></td>
<td>0.75%</td>
<td>1.53*10\textsuperscript{12}</td>
</tr>
<tr>
<td></td>
<td>1%</td>
<td>8.53*10\textsuperscript{11}</td>
</tr>
<tr>
<td>5/95 PG/H\textsubscript{2}O</td>
<td>0.5%</td>
<td>1.57*10\textsuperscript{2}</td>
</tr>
<tr>
<td></td>
<td>0.75%</td>
<td>28.85</td>
</tr>
<tr>
<td></td>
<td>1%</td>
<td>6.35</td>
</tr>
<tr>
<td>15/85 PG/H\textsubscript{2}O</td>
<td>0.5%</td>
<td>0.83</td>
</tr>
<tr>
<td></td>
<td>0.75%</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td>1%</td>
<td>1.9*10\textsuperscript{-3}</td>
</tr>
</tbody>
</table>
Fig. 6.2. Thermal conductivity ratio of DI-H$_2$O based nanofluids w.r.t. time.

The band indicates the period when convection experiments were conducted.

All $k$ results indicate that the nanofluid $k$ equals the M-G model prediction before nanoparticle aggregation starts. For the same nanofluid volume fraction, Figs. 6.2-6.4 also show the peak ratio of nanofluid thermal conductivity ($k/k_b$) is almost identical but has different incident times depending on the base fluid. The different peak times are due to the different $t_p$ of each nanofluid. Almost identical peak ratios are due to the $k_b$ being not strongly affected by increasing PG volume fraction from 0 to 15 vol% in DI-H$_2$O, and hence ratios of $k_p$ to $k_b$ are nearly constant once the nanoparticle loading is set.
Fig. 6.3. Thermal conductivity ratio of 5/95 PG/H₂O based nanofluids. Band indicates the period when convection experiments were conducted.

Fig. 6.4. Thermal conductivity ratio of 15/85 PG/H₂O based nanofluids. Band indicates the period when convection experiments were conducted.
In Fig. 6.2 (DI-H$_2$O based nanofluids), the prediction shows that the nanofluid does not aggregate during the time period for the convection experiments, and thus $k$ is the same as the M-G prediction for fully dispersed nanofluids. For PG/H$_2$O based nanofluid (Figs. 6.3 and 6.4), it is shown during the time period for the convective experiments that there should be some aggregation, meaning that $k$ changes with time and the aggregation stage. In order to determine $k$ within the convective experimental period, an averaged $k$ within the green-band limit (as shown in Fig 6.2 to 6.4) is calculated. Also note that this model does not include any thermal contact resistance between the nanoparticles and the fluid.

6.2. Nu results in thermally developing region

Figures 6.5-6.7 present all the Nu results in the thermally developing region under laminar flow conditions. Figure 6.5 (a)-(c) are results for DI-H$_2$O based nanofluids, Fig. 6.6 (a)-(c) are for 5/95 PG/H$_2$O based nanofluids and Fig. 6.7 (a)-(c) are for 15/85 PG/H$_2$O based nanofluids.

Generally, it is noticed that $Nu$ and $Nu$ enhancement increase with increasing $Re$ and the volume fraction of the nanofluid from Figs. 6.5-6.7. Another general trend is that the $Nu$ enhancement decreases with increasing axial distance from the entrance. It is also observed that not all nanofluids have $Nu$ greater than its base fluid. Among all nanofluids, it is shown that DI-H$_2$O based nanofluids have greatest $Nu$ and $Nu$ enhancement.
Fig. 6.5 (a). $Nu$ vs. $x/L$ of DI-H$_2$O based nanofluids with Al$_2$O$_3$ nanoparticles, for thermally developing flow with $Re \sim 25$.

Fig. 6.5 (b). $Nu$ vs. $x/L$ of DI-H$_2$O based nanofluids with Al$_2$O$_3$ nanoparticles, for thermally developing flow with $Re \sim 120$. 
Fig. 6.5 (c). $Nu$ vs. $x/L$ of DI-H$_2$O based nanofluids with Al$_2$O$_3$ nanoparticles, for thermally developing flow with $Re \sim 210$.

The greatest $Nu$ enhancement of DI-H$_2$O based nanofluids is around 20% and the condition is 1 vol% Al$_2$O$_3$-H$_2$O nanofluid with $Re \sim 210$. When $x/L$ reaches 0.41, the $Nu$ enhancement becomes 6% for the nanofluid. For low nanoparticle loading and low flow rate, such as 0.5 vol% Al$_2$O$_3$-H$_2$O nanofluid with $Re \sim 25$, the $Nu$ results of nanofluids are less than the results of pure DI-H$_2$O.
Fig. 6.6 (a). $Nu$ vs. $x/L$ of 5/95 PG/H$_2$O based nanofluids with Al$_2$O$_3$ nanoparticles, for thermally developing flow with $Re \sim 20$.

Fig. 6.6 (b). $Nu$ vs. $x/L$ of 5/95 PG/H$_2$O based nanofluids with Al$_2$O$_3$ nanoparticles, for thermally developing flow with $Re \sim 110$. 

99
Fig. 6.6 (c). $Nu$ vs. $x/L$ of 5/95 PG/H$_2$O based nanofluids with Al$_2$O$_3$ nanoparticles, for thermally developing flow with $Re \approx 200$.

Fig. 6.7 (a). $Nu$ vs. $x/L$ of 15/85 PG/H$_2$O based nanofluids with Al$_2$O$_3$ nanoparticles, for thermally developing flow with $Re \approx 10$. 
Fig. 6.7 (b). $Nu$ vs. $x/L$ of 15/85 PG/H$_2$O based nanofluids with Al$_2$O$_3$ nanoparticles, for thermally developing flow with $Re \sim 55$.

Fig. 6.7 (c). $Nu$ vs. $x/L$ of 15/85 PG/H$_2$O based nanofluids with Al$_2$O$_3$ nanoparticles, for thermally developing flow with $Re \sim 100$. 
In Figs. 6.6 (a)-(c) and 6.7 (a)-(c), the PG/H$_2$O based nanofluids share the same trends as the DI-H$_2$O based nanofluids. The greatest $Nu$ and $Nu$ enhancement are always at 1 vol% nanofluids and with the highest $Re$. The $Nu$ results of 5/95 PG/H$_2$O based nanofluids are greater than the base fluid in the entrance region except for the 0.5 vol% nanofluid at $Re$~20. The 15/85 PG/H$_2$O based nanofluids exhibit $Nu$ values greater than that for the base fluid only very near the entrance. Besides the $Nu$ at the entrance, all 15/85 PG/H$_2$O based nanofluids have $Nu$ less than the base fluid. Note that the $Re$ approximations were due to the test conditions that were set at controlled volume flow rates.

6.3. $Nu$ results in the fully developed region

The $Nu$ vs. $Re$ results for DI-H$_2$O based nanofluids in the fully developed region are presented in Fig. 6.8. The nanofluids’ results show that $Nu$ of nanofluids increase with increasing $Re$ [46] in fully developed flow region. For 0.5 and 0.75 vol% nanofluids, there is only a small difference of $Nu$ results for different nanofluid volume fraction but the $Nu$ trend clearly increases as $\phi$ increases. In addition, the $Nu$ results for 0.5 and 0.75 vol% nanofluids are less than 4.364, the classical theoretical prediction for constant wall heat flux. The 1 vol% Al$_2$O$_3$ nanofluid has $Nu$ greater than 4.364, albeit at the expense of greater viscosity and pressure drop.
Fig. 6.8. $Nu$ vs. $Re$ of DI-H$_2$O based nanofluids with Al$_2$O$_3$ nanoparticles.

Fig. 6.9. $Nu$ vs. $Re$ of 5/95 PG/H$_2$O based nanofluids with Al$_2$O$_3$ nanoparticles.
Fig. 6.10. $Nu$ vs. $Re$ of 15/85 PG/H$_2$O based nanofluids with Al$_2$O$_3$ nanoparticles.

Figures 6.9 and 6.10 present the $Nu$ vs. $Re$ results for Al$_2$O$_3$ nanofluids in 5/95 and 15/85 PG/H$_2$O mixtures, respectively. The general trend is the same as for the DI-H$_2$O based nanofluids in Fig. 6.8, in that $Nu$ increases with increasing $Re$ and nanoparticle volume fraction. It is shown that with increasing PG volume fraction, $Nu$ decreases and the $Nu$ values of all 15/85 PG/ H$_2$O based nanofluids are below 4.364. Only 1 vol% Al$_2$O$_3$ nanoparticles in the 5/95 PG/ H$_2$O mixture have $Nu$ greater than 4.364 at higher $Re$. The data of 0.5 and 0.75 vol% of PG/H$_2$O based nanofluids do not show much difference, especially at low $Re$. 
6.4. Discussion

Since $Nu$ is inversely proportional to the thermal boundary layer thickness, the observed greater $Nu$ enhancement at the entrance region could still be explained by the thermal boundary layer development. As the thermal boundary layer thickness builds up, the enhancement of $k$ did not seem conclusively to have a positive effect on the $h$ enhancement of nanofluids and hence the $Nu$ of the nanofluid is less than the $Nu$ of the base fluid.

For the fully developed region, theoretically, the $Nu$ value equals a constant if a steady laminar flow of an incompressible fluid with constant properties in a straight circular test tube is considered [61]. The $Nu$ number equals 4.364 while applying constant heat flux at the wall and is 3.66 for a constant wall temperature [61]. Relatively few studies have reported results in nondimensional form for nanofluids, i.e., as $Nu$ versus $Re$. However, for the few studies where the $Nu$ number of nanofluid was reported, a clear increasing trend with increasing $Re$ [35, 39, 46] was usually evident, although the magnitude of the enhancement in $Nu$ was not consistent. Heris et. al [39] reported their $Nu$ increasing with Peclet number ($Pe = RePr$), which indicated their tested $Re$ was within 700 to 2050. Their results show that the $Nu$ of nanofluids was always greater than the theoretical prediction. Yang et. al [35] and Jung et al [46] both reported $Nu$ increased with increasing $Re$ but the $Nu$ values were all much less than the theoretical prediction. This increasing trend of $Nu$ might
have resulted from the higher flow rate mixed with nanoparticle Brownian motion, which intensified the energy exchange rate between tested fluid and the tube wall.

Results also show that only a few cases have $Nu$ greater than 4.364 for laminar flow in a fully developed region while adopting the $k$ model [62] and assuming constant nanofluid properties. At first glance, the $Nu$ of nanofluid decreases when higher viscosity was observed. Under the same nanoparticle volume fraction, the higher the viscosity of the base fluid is, the lower the $Nu$ that resulted. The mechanism that leads to the lack of $h$ enhancement may be due to the undesired orientation of aggregates along with aggregate shapes within the flow field, which thus lowers the heat transfer rate.

It is shown that the nanoparticles exhibit signs of aggregation from DLS size measurements and it is also shown that the time dependent $k$ results indicate that the aggregation stage changes with respect to time and hence changes $k$. Per the model of Prasher et. al. [62], the calculated strong aggregation predicts that a 1 vol% Al$_2$O$_3$ in 15/85 PG/H$_2$O based nanofluid aggregates into a single cluster suspended in the base fluid within 100 minutes. This is directly in conflict with the results of the DLS size measurements and observations during the convection experiments. Hence, in order to include the measured aggregation size, a combined theoretical/experimental approach is introduced hereafter. This modified approach is based on the theoretical model of Prasher et. al. [62] and the
DLS size-measurement results in Chapter 3. From Figs. 3.7-3.8, the aggregate size in 0.5 vol% and 0.75 vol% PG/H$_2$O nanofluids show an increasing trend with respect to time. Therefore, a distribution of aggregation size versus time can be plotted as shown in Fig. 6.11. Please note that only four samples are presented in Fig. 6.11. Other non-presented nanofluid samples had an inconclusive trend on their size measurement results.

Using the measured size as the gyration radius of the aggregates ($R_a$) and applying the eq. (6.6), the volume of nanoparticles to the volume of aggregate, $\varphi_{int}$, can be obtained. With the eq. (6.7) the aggregation time constant $t_p$ can be calculated and is also shown in Fig. 6.11. It is
interesting to show that the calculated $t_p$ of 5/95 PG/H$_2$O based nanofluids by combined theoretical/experimental prediction is significantly shorter than those based on purely theoretical estimations. Hence, it is suggested that, for 5/95 PG/H$_2$O based nanofluids, the nanofluids show more signs of aggregation from the combined experimental/theoretical approach than the purely theoretical prediction [62], and the maximum thermal conductivity enhancement moves forward with respect to time as in Fig. 6.12.

In Fig. 6.12, the band again indicates the period of time when convection experiments were conducted. The purely theoretical results [62] show that the nanofluids were in the early stage of aggregation after one hour. However, with the combined experimental/theoretical method, the nanofluids passed their thermal conductivity enhancement apexes where the $k_p$ was dominant due to aggregation. However, compared with the $k$ results from the purely theoretical prediction, the $k$ results by the modified approach have greater enhancement due to their stage of aggregation.
Fig. 6.12. Comparison of thermal conductivity ratio by different aggregation time constant. Black lines are results by the purely theoretical model of [62] and green lines indicate the results from combined experimental/theoretical predictions. Nanofluids are 0.5 vol% and 0.75 vol% Al₂O₃ in 5/95 PG/H₂O base fluids.
Fig. 6.13. Comparison of thermal conductivity ratio with different aggregation time constant. Black lines are the results by the purely theoretical model of [62] and green lines indicate the results from the combined experimental/theoretical predictions. Nanofluids are 0.5 vol% and 0.75 vol% Al$_2$O$_3$ in 15/85 PG/H$_2$O base fluids.

The $k$ results by the modified [62] model shown in Fig. 6.13 are 15/85 PG/H$_2$O based nanofluids. Compared with the theoretical predictions, the $k$ distribution shows a peak around 30 minutes after ultrasonication for 0.5 vol% 15/85 PG/H$_2$O nanofluid. The results for 0.75 vol% 15/85 PG/H$_2$O nanofluid do not show much difference between the modified and non-modified approaches due to the close $t_p$ between the experimental (DLS) aggregation rate and the predicted aggregation rate.
Fig. 6.14. Comparisons of \( \text{Nu} \) vs. \( \text{Re} \) using modified and non-modified \( k \) of PG/H\(_2\)O based nanofluids. The blue line indicates the constant \( \text{Nu} \) result from the traditional analytical solution. The closed symbols are the \( \text{Nu} \) results when using \( k \) results by the combined experimental/theoretical approach and the open symbols are \( \text{Nu} \) when using \( k \) results by predictions of [62].

In Fig. 6.14, compared with the original \( \text{Nu} \) results from Fig. 6.9 and 6.10, the \( \text{Nu} \) results of 5/95 PG/H\(_2\)O based nanofluids decreased when adopting the modified \( k \) approach. \( \text{Nu} \) results of 15/85 PG/H\(_2\)O based nanofluid have no (0.75 vol% Al\(_2\)O\(_3\) 15/85 PG/H\(_2\)O nanofluid) or slightly increased (0.5 vol% Al\(_2\)O\(_3\) 15/85 PG/H\(_2\)O nanofluid) \( \text{Nu} \) values while adopting the modified \( k \) approach. However, it is also interesting to point
out the change in \( Nu \) results due to different \( k \) approaches is insignificant for 15/85 PG/H\(_2\)O based nanofluids.

As for effects of cluster shape, the nanoparticles appear to have elongated aggregates when the nanofluid is subjected to laminar flow in the test tube with constant heat flux at the wall, as shown in Chapter 4 by application of Brenner’s model [86]. Compared with suspensions containing individual dispersed nanoparticles or relatively spherical clusters, the elongated suspensions could complicate the flow field further based on the aggregates’ orientation. The aggregates not only rotate under the influence of the shear stress of the flow field, but also additionally change the heat transfer direction by its high thermal-conductivity percolation path. Once the high thermal-conductivity percolation path is parallel to the main stream direction, it is likely to lead to increased axial conduction in the flowing nanofluids. In section 5.4, it was hypothesized that the enhancement of \( h \) is due to the \( k \) distribution along the radial direction and the change of velocity profile in test channel. However, the conduction direction was not considered in that section. The significant parameter of axial conduction is the Peclet number (\( Pe \)), which is defined as \( Pe = Re \cdot Pr \). The \( Pe \) ranges from around 150 to 1200 and the effect of fluid axial conduction is usually negligible when \( Pe > 100 \) [61]. However, considering our mm-size test tube, the \( k \) ratio of the nanofluid and test tube and the nanoparticle distribution, the conduction in the axial direction may become a competitive mechanism of heat transfer. For the
fluid axial conduction part, assume the laminar nanofluid flow within the test tube can be treated as a core-annular flow, i.e. as a concentrated core with nearly base fluid outer layer. Su [100] demonstrated that with a decreasing $k$ ratio of outer layer to inner core fluids, that $Nu$ decreases.

As for the wall axial conduction, Morini [101] suggested the following inequality to establish its significance:

$$\frac{k_w D_o - D_i}{k D_i L} \frac{1}{Re Pr} > 0.01$$  \hspace{1cm} (6.10)

Although eq. (6.10) was suggested for microchannels, the criterion holds for mini-sized channels, too [102]. The minimum results of eq. (6.10) of this study is 0.18 which occurs at 1 vol% 15/85 PG/H$_2$O nanofluid with a tested volume flow rate equal to 1 mL/min. Therefore, it is possible that wall axial conduction affects the convective heat transfer results. The study of Sakakibara et al. [103] showed that, when strong axial wall conduction is present, the high axial conduction rate levels out the axial temperature distribution at the wall-fluid interface even when a constant wall heat flux was applied. The greater the $k_w/k$ becomes, the more uniform the temperature along the tube and wall-fluid interface tends to be. Therefore, the $Nu$ results of fully developed laminar move toward 3.66, the constant wall temperature condition, once the axial wall conduction presents.
6.5. Summary

In this chapter, the $Nu$ results for the tested nanofluids under laminar flow conditions were presented. The $k$ of nanofluid was predicted by [62] and its modified approach where the aggregation size of nanoparticles by DLS measurement was adopted. Using the theoretical model of [62] and considering the temperature effects on the base fluid, the $Nu$ results show an increasing trend with increasing $Re$. The possible explanation of this increasing trend could be the higher volume flow rate plus the nanoparticle motion and enhanced energy exchange rate between the fluid and the tube wall. The $Nu$ results of nanofluids also showed that nanofluids have more effective thermal conduction capability than convection ability in fully developed laminar flow condition except 1 vol\% Al$_2$O$_3$ nanoparticles in DI-H$_2$O and 5/95 PG/H$_2$O cases. The combined experimental/theoretical approach, which adopted the nanoparticle size results from DLS measurements, show that the nanofluids had stronger aggregation characteristics than the purely theoretical prediction and the higher thermal conductivity from aggregation lowered the $Nu$ number for 5/95 PG/H$_2$O based nanofluids. On the contrary, for 15/85 PG/H$_2$O based nanofluids, the nanofluids benefited by the aggregation rate and showed a slight increase in $Nu$. 
Chapter 7: Conclusions and future work

When considering convective cooling methods, an ideal coolant should possess substantial characteristics such as high thermal conductivity, high thermal capacity, and low viscosity. Many researchers show that nanofluids have promising potential in several respects, for example extraordinary enhancement in thermal conductivity of base fluids [13-15] and Newtonian behavior [27, 75-79]. The research reported in this dissertation has attempted to answer if nanofluids are a better coolant than their base fluid through several aspects, namely nanofluids' characterization, viscosity and convective heat transfer performance. The conclusions and suggested future works are as follows.

7.1. Conclusions

From the nanofluid characterization results, it is shown that the nanofluid stability is sensitive to the base fluid compounds. For example, a slight change in the volume fraction of two fluids’ mixture can change the Hamaker constant and significantly reduce the stability of a nanofluid. Compared with its base fluid, the calculation of nanofluid thermal conductivity, $k$, also indicates that the aggregation stage of the nanoparticles changes the $k$ enhancement. Therefore, precautions should be taken when selecting materials for nanofluids. The density difference between the nanoparticles and the base fluid should also be considered to avoid any sedimentation.
The viscosity results indicate that the relative viscosity, $\mu_r$, increases with nanoparticle and PG volume fraction. For unheated cases, the data did not show any conclusive trend with respect to nanofluid volume flow rate. For the cases with applied constant heat flux, $\mu_r$ showed decreasing trends with respect to increasing volume flow rate. Nanofluids flowing in heated tubes exhibited higher $\mu_r$, which may be due to highly frequent particle interactions as the nanofluid temperature increases, and possibly to the effect of thermophoretic flow from the hotter downstream end to the cooler upstream end. Using the viscosity model by Brenner [86], the aggregation size of nanoparticle suspensions was calculated and suggested the aggregation had an elongated chain shape. There may be some difference in how the aggregate aspect ratio changes with increasing volume fraction, depending on whether or not the tube is heated.

The heat transfer coefficient result, $h$, of the tested nanofluids showed that nanofluids yield higher $h$ than the base fluids. Results also showed that $h$ increases with volume flow rate and nanoparticle volume fraction. The $h$ enhancement is significant in the entrance region and decreased with increasing axial distance from the test section entrance. These results also demonstrated that the higher the nanoparticle volume fraction, the longer is the thermal entrance length. The $h$ enhancements also increased with increasing volume flow rate and nanoparticle volume fraction. Once the PG volume fraction increased in PG/H$_2$O mixture, the $h$ and $h$ enhancement decreased. The enhancement in $h$ in the entrance
region for nanofluids can be explained, at least in part, by classic boundary layer thickness effects.

The $Nu$ results of nanofluids in the fully developed region increased with increasing $Re$ and nanoparticle loading. The enhancement of $k$ did not seem conclusively to have a positive effect on the $h$ enhancement of nanofluids in this study. The lack of enhancement of $h$ led to only two 1 vol% nanofluids that have $Nu$ greater than the theoretical prediction, 4.364, at flow volume rates at 5 and 9 mL/min in the fully developed region.

Judging by the heat transfer results of the present work, it is suggested that the length of the flow channel when utilizing nanofluid as the heat transfer fluid is better within the developing region rather than the fully developed region. A higher volume flow rate of nanofluid or $Re > 100$ with $\varphi \geq 1$ vol% is also recommended. There is a noticeable reduction in $h$ and $Nu$ when the PG concentration was increased. Therefore, pure deionized water is shown to be the best base fluid among all tested cases.

7.2. Future work

Experimental works in this study have shown some potential improvements by using nanofluids for laminar convective heat transfer. The following questions, however, should be addressed in future work:

1. Per [62], the aggregation stage of nanoparticles in suspension, calculated by DLVO theory or measured by DLS, was considered and thermal conductivity of nanofluid was able to be calculated. However,
the model did not include the effects from the flow field. Therefore, the aggregation kinetics and its related thermal conductivity models in flowing conditions are unknown and require further research.

2. From the data and analysis of nanofluid viscosity, this study showed that nanofluid could have an elongated aggregation chain in the flow field. Therefore, it is of interest to understand how nanoparticles will align in the flow field and affect the convective heat transfer path.

3. Different base fluids and mixtures were studied. It was shown that different base fluids changed the Hamaker constant between nanoparticles and hence changed the adhesive potential between nanoparticles. The thermal conductivity results also indicated the nanoparticle aggregation stage could change the enhancement magnitude of nanofluid thermal conductivity. The change in adhesive potential between particles will also affect aggregation kinetics no matter if the nanofluid is stationary or under flow. Therefore, will different particle combinations affect convective heat transfer efficiency?

4. It is interesting to point out that since the size of nanoparticles is extremely small, the interaction phenomena between nanoparticles, fluid molecules and the tube wall remains unknown. Is there an optimal condition, such as flow loop geometry, nanoparticle size to flow loop diameter, etc., when utilizing specific nanofluids?
5. Through the positive preliminary boiling research of Taylor and Phelan [99], it is interesting to know what is the possible maximum boundary heat flux before the boiling bubble clogs the test tube when utilizing nanofluids for internal convection applications.

6. As for stability point of view, how will nanofluids perform over a long time period?
References


[34] D. Wen, Y. Ding, Experimental investigation into convective heat transfer of nanofluids at the entrance region under laminar flow conditions 47 (2004) 5181-5188.


126


APPENDIX A

MODIFIED HAMAKER CONSTANT OF BINARY LIQUID MIXTURE
The Hamaker constant for two identical phases 1 interacting across a medium 3 is given in [97].

\[
A_{131} = \frac{3}{4} k_B T \left( \frac{\varepsilon_1 - \varepsilon_3}{\varepsilon_1 + \varepsilon_3} \right)^2 + \frac{3\hbar v_e}{16\sqrt{2}} \frac{\left( n_1^2 - n_3^2 \right)^2}{\left( n_1^2 + n_3^2 \right)^{3/2}}
\]  

(A1)

The \(k_B\) is the Boltzmann constant, \(T\) is the temperature, \(\hbar\) is Plank constant, \(v_e\) is the plasma frequency of the free electron gas and typically around \(3 \times 10^{15} \text{ s}^{-1}\), \(\varepsilon\) is the dielectric permittivity of a medium and \(n\) is the refractive index of the medium in the visible.

The \(n\) and \(\varepsilon\) of a mixture of two transparent liquids can be calculated by [97]

\[
n = n_{SL} + \phi_{sl} \left( n_{SL} - n_{sl} \right)
\]

(A2)

\[
\varepsilon = \varepsilon_{SL} \left( 1 - \phi_{sl} \right) + \phi_{sl} \varepsilon_{sl}
\]

(A3)

The subscript \(SL\) denotes solvent and \(sl\) is for solute.