NUMERICAL MODELLING OF BUBBLY FLOWS IN NANOFLOUIDS WITH AND WITHOUT HEAT TRANSFER

A thesis submitted in fulfilment of the requirements for the degree of Doctor of Philosophy

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Declaration

I certify that except where due acknowledgement has been made, the work is that of the author alone; the work has not been submitted previously, in whole or in part, to qualify for any other academic award; the content of the thesis is the result of work which has been carried out since the official commencement data of the approval research program; and, any editorial work, paid or unpaid, carried out by a third party is acknowledged; and, ethics procedures and guidelines have been followed.

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Abstract

Nanofluids are engineered colloidal dispersions of nano-scale particles (nanoparticles hereafter) in water, or other base liquids. This thesis focuses on the bubbly flows in nanofluids with and without heat transfer. For the former, the nucleate boiling of dilute nanofluids (≤0.01 vol%) in cylindrical containers are investigated numerically. For the latter, the two-phase flows of dilute nanofluids in vertical tubes are numerically studied.

Dilute nanofluids exhibits largely improved heat transfer performances during both pool and flow boiling, whilst being compared with corresponding pure liquid, and these properties make nanofluid suitable as a heat transfer medium in a stream of equipment dealing with extremely high heat flux and needing high cooling efficiency. Despite the many advantages, the use of nanofluid in industry is still limited. Two major research gaps remain between the preliminary studies and industry applications. One is the difficulty to accurately describe the boiling heat transfer and efficiently predict the relevant heat transfer coefficient (HTC). Because of the inherent complexity, this requires an in-depth understanding of the heated surface characteristics and bubble hydrodynamics in the near-wall region, for both pool and flow boiling. Beyond that, for flow boiling of nanofluids, the heat transfer is closely related to the two-phase flow structures, which needs particular attention. However, to the best of the author’s knowledge, relevant numerical and mechanistic studies are still absent in the open literature. The lack of studies in two-phase flow structures and dynamics is another gap which makes the prospect of nanofluid’s application in industry much gloomier. Recently, with rapid development of computer technology and computational algorithm, Computational Fluid Dynamics (CFD) provides a powerful numerical approach to conduct simulation on gas-nanofluid bubbly flows, and further explore the underlying mechanism behind.

The main body of this thesis is composed of four parts. In the first part (Chapter 2), a comprehensive literature review, including fundamentals of pool and flow boiling, experimental studies of dilute nanofluids and preliminary numerical modelling of two-phase gas-liquid bubbly flows, was performed to identify the research gaps between
previous studies and numerical modelling of dilute nanofluids. In the second part (Chapter 3), a parametric study of the heat flux partitioning (HFP) model for nucleate boiling of nanofluids was conducted with the consideration of the effects of nanoparticle deposition on the heated surface characteristics and bubble behaviours in the near-wall region. Moreover, a new HFP model was proposed, in which a new heat flux component was incorporated to account for the heat transfer by the nanoparticle Brownian motion in microlayer. In the third part (Chapter 4), the flow structures and dynamics of two-phase flows of dilute nanofluids were investigated with the two-fluid model and MUtiple-SIze-Group (MUSIG) model, respectively. In order to identify the individual factors affecting the hydrodynamic behaviours, the heat transfer was not considered. The simulation results showed that both of the above two models need substantial improvement in order to achieve an effective modelling of nanofluids. In the fourth part (Chapter 5), mechanistic studies on the role that nanoparticles have played in affecting the bubble-liquid and bubble-bubble interactions were conducted to clarify the theoretical frame which could be used to develop predictive models for two-phase gas-liquid flows containing nanoparticles.

In summary, the effects of nanoparticles on boiling heat transfer and flow structures in gas-nanofluid bubbly flows were investigated with and without heat transfer, respectively, and the preliminary heat flux partitioning (HFP) model, two-fluid model as well as the MUSIG model were further developed accordingly. Numerical results were compared with experimental data, which validated the feasibility of new models in simulating nanofluids.
# Contents

1 Introduction .......................................................................................................................... 1

1.1 Background and Motivation ............................................................................................. 1

1.1.1 Thermo-physical Properties ....................................................................................... 2

1.1.2 Single-phase Convective Heat Transfer ..................................................................... 3

1.1.3 Boiling Heat Transfer .................................................................................................. 4

1.2 Objectives ......................................................................................................................... 6

1.3 Thesis Outline ................................................................................................................... 7

2 Literature Review ................................................................................................................ 9

2.1 Overview of Boiling Heat Transfer .................................................................................... 9

2.1.1 Pool Boiling .................................................................................................................. 9

2.1.2 Flow Boiling ............................................................................................................... 12

2.2 Experimental Findings of Nucleate Boiling of Nanofluids ............................................ 14

2.2.1 Pool Boiling Heat Transfer ....................................................................................... 14

2.2.1.1 Enhancement ......................................................................................................... 16

2.2.1.2 Deterioration ......................................................................................................... 17

2.2.1.3 Both Enhancement/Deterioration ........................................................................ 18

2.2.2 Influencing Factors ..................................................................................................... 19

2.2.2.1 Thermo-physical Properties .................................................................................. 20

2.2.2.2 Characteristics of the Heated Surface .................................................................... 22

2.2.2.3 Near Surface Hydrodynamics ............................................................................... 28

2.2.2.4 Bulk Field Hydrodynamics .................................................................................. 34

2.3 Numerical Modelling of Gas-liquid Flows .................................................................... 37

2.3.1 Numerical Modelling of Boiling Heat Transfer ....................................................... 37

2.3.1.1 Heat Flux Components ......................................................................................... 37

2.3.1.2 Boiling Parameters ............................................................................................... 38

2.3.2 Numerical Modelling of Bulk Flow .......................................................................... 41

2.3.2.1 Two-fluid Model .................................................................................................. 41
2.3.2.2 MUtiple-SIze-Group (MUSIG) Model ............................................. 45
2.3.3 Main Challenges in Modelling Bubbly Systems of Nanofluids ................. 49
3 Numerical Modelling of Boiling Heat Transfer in Dilute Nanofluids ............... 51
3.1 A Parametric Study of the Heat Flux Partitioning Model for Nucleate Boiling of Nanofluids ................................................................. 52
  3.1.1 Introduction .................................................................................. 52
  3.1.2 The Heat Flux Partitioning (HFP) Model .................................... 54
    3.1.2.1 The Active Site Density ...................................................... 55
    3.1.2.2 Other Nucleate Boiling Parameters .................................... 59
  3.1.3 Numerical procedures ................................................................. 61
  3.1.4 Results and Discussion ................................................................ 63
    3.1.4.1 Comparison against experimental data ................................ 63
    3.1.4.2 Prediction of pool boiling experimental data of aqueous-oxide nanofluids ................................................................................ 66
  3.1.5 Conclusions .................................................................................. 70
3.2 A Theoretical Model Considering the Nanoparticle Brownian Motion in Liquid Microlayer ................................................................. 72
  3.2.1 Introduction .................................................................................. 72
  3.2.2 Heat Flux Partitioning in Nucleate Boiling of Nanofluids ................. 75
    3.2.2.1 Heat Flux Partitioning in Boiling Nanofluids ....................... 75
    3.2.2.2 Heat Transfer by Nanoparticle Brownian Motion in the Microlayer .... 78
  3.2.3 Results and Discussion ................................................................ 84
    3.2.3.1 Model Validation and Analysis of HFP Components .......... 84
    3.2.3.2 Analyses of the Influencing Parameters .............................. 88
  3.2.4 Conclusions .................................................................................. 93
4 Numerical Modelling of Two-phase Flows of Dilute Nanofluids .................. 94
4.1 Two-fluid Modelling of Air-nanofluid Bubbly Flows .............................. 95
  4.1.1 Introduction ................................................................................ 95
  4.1.2 Modelling of Bubbly Flow in a Vertical Tube .............................. 98
4.1.2.1 The Two-fluid Model ................................................................. 98
4.1.2.2 Numerical Procedures ............................................................... 100
4.1.3 Results and Discussion ................................................................ 101
  4.1.3.1 Model Applicability to Water and Nanofluid .......................... 101
  4.1.3.2 Model Improvement for Air-nanofluid Bubbly Flows ............... 106
  4.1.3.3 Effects of Nanoparticles on the Interfacial Behaviours ............ 110
4.1.4 Conclusions .................................................................................. 112

4.2 Multi-particle-Sized-Group (MUSIG) Modelling of Air-nanofluid Bubbly Flows in a Vertical Tube .......................................................... 114
  4.2.1 Introduction ................................................................................. 114
  4.2.2 The MUSIG Model .................................................................... 116
    4.2.2.1 The Flow Equations ............................................................... 116
    4.2.2.2 Population Balance Method .................................................... 118
  4.2.3 Numerical Procedure ................................................................. 121
  4.2.4 Results and Discussion ............................................................... 123
    4.2.4.1 Comparison of simulation results against experimental data ...... 123
    4.2.4.2 Model Improvement for the effects of nanoparticle self-assembly .... 125
    4.2.4.3 Effects of Nanoparticle Self-assembly on Liquid Film Drainage .... 133
  4.2.5 Conclusions .................................................................................. 137

5 Mechanistic Study of Bubble Hydrodynamics in Nanofluids ............... 138
  5.1 Mechanistic Analysis of the Effects of Nanoparticles on Interfacial Forces on Bubbles in Nanofluids ................................................................. 139
    5.1.1 Introduction ................................................................................. 139
    5.1.2 Theoretical Models ................................................................. 141
    5.1.3 Results and Discussion ............................................................... 145
      5.1.3.1 Comparison of the Numerical Results against Experimental Data .... 145
      5.1.3.2 The adsorption of nanoparticles on air-water interface .......... 146
      5.1.3.3 Analysis of the Lift Force ....................................................... 148
      5.1.3.4 Analysis of the Drag Force ..................................................... 153
      5.1.3.5 Summary and Key Research Points ....................................... 155
5.1.4 Conclusions ........................................................................................................ 157
5.2 Effects of Spontaneous Nanoparticle Adsorption on the Bubble-liquid and Bubble-bubble Interaction ........................................................................................................ 158
  5.2.1 Introduction ........................................................................................................ 158
  5.2.2 Nanoparticle Adsorption at Phase Interfaces .................................................. 160
  5.2.3 The Influences of Nanoparticles on Bubble-liquid Interactions ................. 162
    5.2.3.1 Bubble-liquid Interaction .......................................................................... 162
    5.2.3.2 The Lift Force ......................................................................................... 164
    5.2.3.3 The Drag Force ....................................................................................... 171
  5.2.4 The Influences of Nanoparticles on Bubble-bubble Interactions ............ 176
    5.2.4.1 Bubble-bubble Interaction ...................................................................... 176
    5.2.4.2 Thinning Process ..................................................................................... 179
    5.2.4.3 Rupture Process ..................................................................................... 186
  5.2.5 Summary .......................................................................................................... 189
6 Conclusions .............................................................................................................. 190
Bibliography .............................................................................................................. 194
List of Figures

Figure 2. 1: Electrical and thermal heating (Naterer, 2002) ................................................................. 10
Figure 2. 2: Nukiyama’s boiling curve (Nukiyama, 1966) ............................................................. 10
Figure 2. 3: Bubble grow and departure on an active site (Li et al., 2014a) ...................................... 11
Figure 2. 4: Flow patterns in vertical upflow: (a) bubbly flow; (b) slug flow; (c) churn flow; (d) annular flow. ......................................................................................................................... 13
Figure 2. 5: Two-phase flow regimes in vertical pipe flow (Naterer, 2002). .......................... 14
Figure 2. 6: Boiling curve of nanofluids on: (a) smooth heater surface ($R_a=0.4\mu m$); (b) roughened heater surface ($R_a=1.15\mu m$) (Das et al., 2003a) .................................................. 15
Figure 2. 7: Boiling curve of pure water and Al$_2$O$_3$/water nanofluids (0.001g/l to 0.05g/l) (You et al., 2003) ......................................................................................................................... 16
Figure 2. 8: Comparative boiling experiments on the smooth surface (Wen et al., 2011) ............................................................................................................................................... 17
Figure 2. 9: Thermal conductivity enhancement of nanofluids as a function of temperature (Das et al., 2003a) ................................................................................................................................. 21
Figure 2. 10: Surface roughness of the smoother heater surface: (a) before boiling; (b) after boiling with nanofluids (Das et al., 2003a). ......................................................................................... 22
Figure 2. 11: Surface roughness of: (a) clear heater ($R_a=37.2$ nm); (b) heater submerged in 0.5 vol% alumina nanofluids ($R_a=67.6$ nm); (c) in 4 vol% alumina nanofluid ($R_a=227.7$ nm) (Bang and Chang, 2005) ......................................................................................................................... 23
Figure 2. 12: Nanoparticle-coated heaters generated by pool boiling experiments of 0.01 vol% nanofluids: (a) TiO$_2$ nanoparticle-coated NiCr wire; (b) Al$_2$O$_3$ nanoparticle-coated NiCr wire; (c) TiO$_2$ nanoparticle-coated Ti wire (Kim et al., 2006a). ........................................ 24
Figure 2. 13: Scanning electron microscope images of stainless steel surface boiling in: (a) pure water; (b) 0.01 vol% Al$_2$O$_3$ nanofluid; (c) 0.01 vol% ZrO$_2$ nanofluid; and (d) 0.01 vol% SiO$_2$ nanofluid (Kim et al., 2006b) ............................................................................................................................................. 24
Figure 2. 14: On surface boiled in pure water: (a) pure water droplet; (b) 0.01 vol% Al$_2$O$_3$ nanofluid droplet; on surface boiled in 0.01 vol% Al$_2$O$_3$ nanofluid: (c) pure water droplet; and (d) 0.01 vol% Al$_2$O$_3$ nanofluid droplet (Kim et al., 2006b) .......................................... 24
Figure 2. 15: Photograph of pool boiling of pure water at 1900 kW/m$^2$ (CHF) on a TiO$_2$ nanoparticle-coated wire with 0.01 vol% nanoparticle concentration (Kim and Kim, 2009) ............................................................................................................................................. 25
Figure 2. 16: Dependency of the maximum capillary wicking height of TiO$_2$ nanoparticle-coated wires on the particle concentration (Kim and Kim, 2009) ................................................... 26
Figure 2. 17: Boiling curves of pure water on nanoparticle-deposited surfaces (Ahmed and Hamed, 2012) ............................................................................................................................................. 27
Figure 2. 18: Effect of surface roughness and particle size on boiling heat transfer (Narayan et al., 2007) ............................................................................................................................................. 27
Figure 2.19: Effects of the surface wettability on the heat transfer coefficient (Phan et al., 2009).

Figure 2.20: Nucleate boiling of pure water (left) and 0.01 vol% Al$_2$O$_3$ nanofluid (right) at the same heat flux on an electrically heated 0.25 mm diameter stainless steel wire (Kim et al., 2006b).

Figure 2.21: Active nucleation site density versus heat flux for contact angles from 18° to 90° (Wang and Dhir, 1993).

Figure 2.22: High speed camera images of a boiling bubble and corresponding liquid-vapour phase boundary, temperature, and heat flux distribution at the boiling surface in nanofluids (Jung and Kim, 2014).

Figure 2.23: Time evolution of the microlayer geometry beneath a growing bubble (Jung and Kim, 2014).

Figure 2.24: Bubble geometries including the microlayer and dry spot during the bubble growth period (Jung and Kim, 2014).

Figure 2.25: Evolution of grow time as a function of contact angle (Phan et al., 2009).

Figure 2.26: Bubbles departing from the wire heater immersed in: (a) pure water; (b) Al$_2$O$_3$/water nanofluid (0.025 g/l) (You et al., 2003).

Figure 2.27: Bubble departure on heater surfaces with various wettability (Phan et al., 2009).

Figure 2.28: Bubble departure frequency versus contact angle (Phan et al., 2009).

Figure 2.29: Average full-field velocity profile for pool boiling of: (a) pure water; (b) Al$_2$O$_3$/water nanofluid (0.002 vol%) (Domínguez-Ontiveros et al., 2010).

Figure 2.30: Effect of (a) nanoparticle concentration; and (b) heat flux on void fraction (Rana et al., 2014).

Figure 2.31: Comparisons of the flow pattern transitions among nitrogen-nanofluid, nitrogen-water/SDBS mixture and nitrogen-water (Wang and Bao, 2009).

Figure 2.32: Comparison of the local two-phase flow parameters: (a) void fraction; (b) bubble velocity; (d) IAC; and (d) mean bubble diameter in the bubbly flow regime (Park and Chang, 2011).

Figure 3.1: Comparison of the Ganapathy-Sajith correlation (Ganapathy and Sajith, 2013) against experimental data (Gerardi et al., 2011): (a) effect of the liquid contact angle; (b) effect of nanoparticle size.

Figure 3.2: The computational domain.

Figure 3.3: Comparison of active site density prediction against experimental data (Gerardi et al., 2011).

Figure 3.4: Bubble departure diameter as a function of the wall superheat.

Figure 3.5: Comparison of bubble departure correlations against experimental data (Gerardi et al., 2011).
Figure 3.6: Predicted wall superheat vs. experimental data (Gerardi et al., 2011) ....... 66
Figure 3.7: Comparison of predicted boiling curves against experimental data .......... 67
Figure 3.8: Effects of liquid contact angle, particle size and nanoparticle material on bubble nucleation ............................................................................................................ 70
Figure 3.9: Nanoparticle concentrating in microlayer as bubble grows .................... 74
Figure 3.10: Bubble departure diameter as a function of the wall superheat .............. 77
Figure 3.11: Evolution of the microlayer sizes as the bubble grows (Jung and Kim, 2014) ......................................................................................................................................................... 79
Figure 3.12: Linear reduction of the microlayer thickness as bubble grows (Jung and Kim, 2014): (a) movement of the microlayer surface; (b) reduction of the microlayer thickness ........................................................................................................................................ 81
Figure 3.13: Nanoparticle concentration in microlayer: (a) evolution of nanoparticle concentration in the microlayer; (b) mean nanoparticle concentration in microlayer vs. the bulk concentration .............................................................................................................. 83
Figure 3.14: The equivalent thermal conductivity of nanoparticle Brownian motion. 84
Figure 3.15: Prediction of the active site density .......................................................... 85
Figure 3.16: Comparison of predicted pool boiling curves against the experimental data (Gerardi et al., 2011) ........................................................................................................................................ 86
Figure 3.17: Comparison of heat flux components by the models: (a) classic HFP model; (b) new HFP model ............................................................................................................ 87
Figure 3.18: Microlayer parameters vs. heat flux ......................................................... 88
Figure 3.19: Effects of the bulk concentration. (Note: SiO$_2$/water, nanoparticle size 34 nm, surface roughness 100 nm) ................................................................. 90
Figure 3.20: Effects of the nanoparticle size. (Note: 0.1 vol% SiO$_2$/water, surface roughness 100 nm) ................................................................................................................................. 90
Figure 3.21: Effects of the nanoparticle material. (Note: 0.1 vol% nanofluids, nanoparticle size 34 nm, surface roughness 100 nm) ................................................................. 91
Figure 3.22: Effects of the nanoparticle material on the quenching and evaporation heat flux components ................................................................................................. 92

Figure 4.1: The computational domain and boundary conditions ................................ 101
Figure 4.2: Comparison the classic two-fluid model against the experimental data of water: (a) void fraction; (b) bubble velocity (Park and Chang, 2011) ...................... 103
Figure 4.3: Comparison the classic two-fluid model against the experimental data of nanofluid: (a) void fraction; (b) bubble velocity (Park and Chang, 2011) .......... 104
Figure 4.4: Prediction of the void fraction development along the tube using the TFM. Note: Due to the large length-to-diameter ratio of the computational domain, the void fraction contours were not shown in actual proportion. ........................................... 105
Figure 4. 5: Comparison of the Ishii-Zuber model (Ishii and Zuber, 1979) and Grace model (Grace and Weber, 1982) for drag force modelling......................................................... 107
Figure 4. 6: The drag coefficient calculated by the Ishii-Zuber model (Ishii and Zuber, 1979). ...................................................................................................................... 107
Figure 4. 7: The lift coefficient changes as a function of bubble size. ................ 108
Figure 4. 8: The two-fluid model with different values $C_L$ values for the air-nanofluid bubbly flow: (a) void fraction; (b) bubble velocity ................................................................. 109
Figure 4. 9: Fluorescence confocal microscope image of water droplets dispersed in toluence, covered with CdSe nanoparticles (Lin et al., 2005) ........................................... 111
Figure 4. 10: The computational domain ................................................................................................................. 122
Figure 4. 11: Comparison of predicted flow parameters against experimental data of the air-water bubbly flow: (a) void fraction; (b) gas velocity; (c) IAC; (d) Sauter mean bubble diameter (Park and Chang, 2011). ......................................................... 124
Figure 4. 12: Comparison of predicted flow parameters against experimental data of the air-nanofluid bubbly flow: (a) void fraction; (b) gas velocity; (c) IAC; (d) Sauter mean bubble diameter (Park and Chang, 2011). ......................................................... 125
Figure 4. 13: Transmission Electron Microscopy (TEM) image of air bubbles surrounded by MAGSILICA@ H8 nanoparticles ($C_p$=20 mg/mL) in ethanol/water mixture (Rodrigues et al., 2011). ..................................................................................................................... 126
Figure 4. 14: The effect of contaminants: (a) ultra-pure liquid with free-slip boundary condition; (b) slightly contaminated liquid with a limited circulation inside the bubble; (c) fully contaminated bubble with no-slip boundary condition (Dijkhuizen et al., 2010a). .................................................................................................................... 127
Figure 4. 15: Comparison of predicted drag coefficients ($\sigma$=0.065 N/m, $\alpha_g$=0.1). ..... 128
Figure 4. 16: Comparison of predicted flow parameters against experimental data of the air-nanofluid bubbly flow: (a) void fraction; (b) gas velocity (Park and Chang, 2011). ......................................................................................................................... 128
Figure 4. 17: Predicted void fraction of the air-nanofluid bubbly flow with $C_L$=-0.025. ........................................................................................................................................ 129
Figure 4. 18: Comparison of predicted lift coefficients with different correlations of bubble aspect ratio. .............................................................................................................. 130
Figure 4. 19: Predicted film drainage time of equal size bubbles ($\epsilon$=0.65 m$^2$/s$^3$)...... 131
Figure 4. 20: Predicted collision efficiency of equal size bubbles ($\epsilon$=0.65 m$^2$/s$^3$) .... 132
Figure 4. 21: Comparison of predicted void fraction against experimental data of the air-nanofluid bubbly flow (Park and Chang, 2011). ......................................................... 133
Figure 4. 22: Comparison of predicted bubble size fraction when $k_d$ take the value of $k_d$ =1.0~2.0. .......................................................................................................................... 133
Figure 4. 23: The surface tension gradient along the radial dimension of the liquid film. ................................................................................................................................. 135
Figure 4.24: The electrostatic double layer force between two negative-charged bubbles.

Figure 5.1: Comparison of predicted bubble velocity and void fraction profile against experimental data: (a) air-water bubbly flow; (b) air-nanofluid bubbly flow (Park and Chang, 2011).

Figure 5.2: TEM image of air bubbles with MAGSILICA@ H8 nanoparticles ($C_p=10\text{mg/mL}$) in ethanol/water mixtures (Rodrigues et al., 2011).

Figure 5.3: The effect of contaminants: (a) ultra-pure liquid with free-slip boundary condition; (b) slightly contaminated liquid with a limited circulation inside the bubble; (c) fully contaminated bubble with no-slip boundary condition (Dijkhuizen et al., 2010a).

Figure 5.4: Lift force on a spherical bubble in pure liquids.

Figure 5.5: Lift forces on a deformed bubble in pure liquids.

Figure 5.6: Lift forces on a nanoparticle-covered spherical bubble in nanofluids.

Figure 5.7: Bubble lift coefficient versus bubble diameter.

Figure 5.8: Predicted bubble velocity and void fraction profile of air-nanofluid bubbly flows with $C_L=-0.025$: (a) Void fraction; (b) Bubble velocity.

Figure 5.9: Comparison of predicted bubble velocity profiles using different drag correlations.

Figure 5.10: Bubble drag coefficient versus bubble Reynolds number.

Figure 5.11: (a) TEM image of air bubbles with MAGSILICA® H8 nanoparticles in ethanol/water mixture (Rodrigues et al., 2011); (b) Fluorescence confocal microscope image of the adsorbed CdSe nanoparticles at toluene/water interface (Lin et al., 2005).

Figure 5.12: Series of TEM images of 6 nm nanoparticle adsorption to the toluene/water interface in different adsorption steps: (a) step 1; (b) step 2; (c) step 3 (Böker et al., 2007).

Figure 5.13: Flow field surrounding the bubble: (a) spherical bubbles in pure liquid; (b) distorted bubbles in pure liquid; (c) spherical bubbles in nanoparticle-containing system.

Figure 5.14: The lift force acting on: (a) spherical bubbles in pure liquid; (b) distorted bubbles in pure liquid.

Figure 5.15: The predicted lift coefficient as a function of bubble diameter (Yuan et al., 2017).

Figure 5.16: Comparison of predicted flow parameters against experimental data of bubbly flows containing nanoparticles with: (a) Tomiyama model (Equation 5.21); (b) $C_L=-0.025$ (Yuan et al., 2017).
Figure 5. 17: Contributions of pressure $C_{L,p}$ and viscous stress $C_{L,v}$ to the total lift coefficient acting on: (a) a contaminated bubble (Fukuta, Takagi et al., 2008); (b) a rigid sphere (Kurose and Komori, 1999). ................................................................. 168
Figure 5. 18: The lift force acting on spherical bubbles in nanoparticle-containing system. .............................................................................................................................................. 171
Figure 5. 19: The predicted drag coefficient as a function of bubble Reynolds number with Ishii-Zuber model (Ishii and Zuber, 1979). ............................................................................. 172
Figure 5. 20: The effect of contaminants: (a) ultra-pure liquid with free-slip boundary condition; (b) slightly contaminated liquid with a limited circulation inside the bubble; (c) fully contaminated bubble with no-slip boundary condition (Dijkhuizen et al., 2010a). ................................................................................................................................................ 173
Figure 5. 21: Comparison of predicted bubble velocity against experimental data of bubbly flows containing nanoparticles with different drag models (Yuan et al., 2017). .................................................................................................................................................. 174
Figure 5. 22: The predicted drag coefficient as a function of bubble Reynolds number with different drag models (Yuan et al., 2017). ........................................................................................................ 176
Figure 5. 23: Schematic overview of the coalescence process of two bubbles. .......... 177
Figure 5. 24: Comparison of predicted void fraction against experimental data of the bubbly flows containing nanoparticles (Yuan et al., 2016). .................................................. 179
Figure 5. 25: Comparison of predicted bubble size fraction when $k_d$ takes the value of $k_d$=1.0-2.0 (Yuan et al., 2016). .............................................................................................................. 179
Figure 5. 26: Drainage of a liquid film under capillary pressure (Rio and Biance, 2014). .............................................................................................................................................................................. 180
Figure 5. 27: The velocity profile of the liquid in the film with: (a) fully mobile interfaces; (b) partially mobile interface; (c) fully immobile interfaces (Liao and Lucas, 2010). .................................................................................................................. 181
Figure 5. 28: The geometry of the liquid film: (a) deformable surfaces; (b) non-deformable surfaces (Liao and Lucas, 2010). ........................................................................................................... 183
Figure 5. 29: Schematic overview of the liquid film with particles residing in (Hunter et al., 2008). ............................................................................................................................................................ 183
Figure 5. 30: Electrostatic double layer force between two nanoparticle-adsorbed bubble interfaces. ................................................................................................................................. 185
Figure 5. 31: Corrugations of bubble interfaces: (a) Without the adsorption of nanoparticles; (b) With the adsorption of nanoparticles (Rio and Biance, 2014)........... 187
List of Tables

Table 2. 1 Comparison of thermo-physical properties between water and dilute nanofluids (Kim, 2009) .......................................................... 21
Table 2. 2 Static contact angle for water and nanofluids on clean and fouled surfaces (Kim et al., 2007) .......................................................... 25

Table 3. 1 Physical properties of the nanoparticle materials and water. .................. 69
Table 3. 2 Liquid contact angle on heater surfaces boiled in different nanofluids (Kim et al., 2007). .......................................................... 69
Table 3. 3 Physical Properties of the nanoparticle materials and water (webbook.nist.gov). .......................................................... 91

Table 5. 1 Employed physical properties for mathematical modelling .................. 144
## Nomenclature

- \( A_x \) : Heater surface area fraction subjected to convection
- \( A_{sg} \) : Interfacial area per unit volume
- \( A_q \) : Heater surface area fraction subjected to quenching
- \( B \) : Body force
- \( B_B \) : Birth rate of bubble number density due to breakage
- \( B_C \) : Birth rate of bubble number density due to coalescence
- \( C_D \) : Drag coefficient
- \( C_L \) : Lift coefficient
- \( C_{TD} \) : Turbulent dispersion coefficient
- \( C_{w1}, C_{w2} \) : Lubrication coefficient
- \( c \) : Solute concentration
- \( c_f \) : Increase coefficient of surface area
- \( c_{pl} \) : Liquid specific heat
- \( D_B \) : Death rate of bubble number density due to breakage
- \( D_C \) : Death rate of bubble number density due to coalescence
- \( d_p \) : Sauter mean bubble diameter
- \( d_{bw} \) : Bubble departure diameter
- \( d_{cr} \) : Critical diameter
- \( d_H \) : Maximum bubble horizontal dimension
\( d_i \)  Bubble diameter of the \( i^{th} \) group

\( d_{np} \)  Nanoparticle diameter

\( E \)  Bubble aspect ratio

\( Eo \)  Eötvös number

\( Eo^* \)  Modified Eötvös number

\( F_D \)  Drag force

\( F_{gl}, F_{lg} \)  Interfacial force

\( F_L \)  Lift force

\( F_{TD} \)  Turbulent dispersion force

\( F_W \)  Wall lubrication force

\( f \)  Bubble departure frequency

\( f_i \)  MUSIG volume fraction of the \( i^{th} \) group bubbles, (dimensionless)

\( g \)  Gravitational acceleration

\( h \)  Liquid film thickness

\( h_c \)  Convective heat transfer coefficient

\( h_0 \)  Initial film thickness

\( h_f \)  Critical film thickness

\( h_{gs} \)  Latent heat of vaporization

\( h_{lg} \)  Inter-phase heat transfer coefficient

\( k_d \)  Empirical constant in the bubble drainage time calculation

\( k_s \)  Empirical constant in the drag coefficient calculation
\( L \)  
Thickness of the polymer layer

\( L_c \)  
Capillary wicking height

\( N \)  
Potential nucleation site density

\( N_a \)  
Active nucleation site density

\( n_i \)  
Bubble number density of the \( i^{th} \) group

\( n_w \)  
Outward vector normal to the wall surface

\( p \)  
The system pressure

\( q \)  
Total heat flux

\( q_{bm} \)  
Heat flux due to nanoparticle Brownian motion

\( q_c \)  
Heat flux due to convection

\( q_e \)  
Heat flux due to evaporation

\( q_{\text{max}} \)  
Critical heat flux

\( q_q \)  
Heat flux due to quenching

\( R_a \)  
Average surface roughness

\( Re_b \)  
Bubble Reynolds number, (dimensionless)

\( R' \)  
Ideal gas constant

\( r_b \)  
Bubble radius

\( r_i \)  
Bubble radius of the \( i^{th} \) group

\( r_{ij} \)  
Equivalent bubble radius of the \( i^{th} \) group and \( j^{th} \) group

\( S_i \)  
Mass variation rate

\( S_{ij} \)  
Bubble number density variation rate
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$s$</td>
<td>Mean distance between the attachment points</td>
</tr>
<tr>
<td>$T$</td>
<td>Temperature</td>
</tr>
<tr>
<td>$T_i$</td>
<td>Liquid temperature in the cell immediately next to the wall</td>
</tr>
<tr>
<td>$T_{sat}$</td>
<td>Liquid saturation temperature</td>
</tr>
<tr>
<td>$T_w$</td>
<td>Wall temperature</td>
</tr>
<tr>
<td>$\Delta T_{sub}$</td>
<td>Liquid subcooling, $T_{sat} - T_i$</td>
</tr>
<tr>
<td>$\Delta T_{sup}$</td>
<td>Wall superheat $T_w - T_{sat}$</td>
</tr>
<tr>
<td>$t_{ij}$</td>
<td>Bubble drainage time</td>
</tr>
<tr>
<td>$t_w$</td>
<td>Bubble waiting time</td>
</tr>
<tr>
<td>$\bar{U}$</td>
<td>Velocity</td>
</tr>
<tr>
<td>$U_T$</td>
<td>Terminal velocity</td>
</tr>
<tr>
<td>$u_s$</td>
<td>Slip velocity</td>
</tr>
<tr>
<td>$u_T$</td>
<td>Turbulent velocity</td>
</tr>
<tr>
<td>$v_i$</td>
<td>Mean volume of the $i^{th}$ group bubbles</td>
</tr>
<tr>
<td>$y_w$</td>
<td>Adjacent point normal to the wall surface</td>
</tr>
</tbody>
</table>

**Greek symbols**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha$</td>
<td>Void fraction, (dimensionless)</td>
</tr>
<tr>
<td>$\Gamma_i$</td>
<td>Mass variation rate of the $i^{th}$ group bubbles due to coalescence</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>Reduced surface potential</td>
</tr>
<tr>
<td>$\delta$</td>
<td>Separation between the surfaces</td>
</tr>
<tr>
<td>$\varepsilon$</td>
<td>Turbulent kinetic energy dissipation</td>
</tr>
</tbody>
</table>
\( \theta \) Bubble collision frequency
\( \theta \) Surface contact angle
\( \theta_{ij}^{LS} \) Bubble coalescence frequency due to laminar shear
\( \theta_{ij}^{T} \) Bubble coalescence frequency due to turbulence
\( \theta_{ij}^{WE} \) Bubble coalescence frequency due to wake entrainment
\( \kappa \) Debye screening length
\( \lambda \) Bubble collision efficiency
\( \mu \) Viscosity
\( \Pi_{c} \) Capillary pressure
\( \Pi_{e} \) Electrostatic double layer force
\( \Pi_{s} \) Steric repulsion force
\( \rho \) Density
\( \rho_{\infty} \) Number density of ion in the bulk solution
\( \sigma \) Surface tension
\( \tau_{ij} \) Bubble contact time
\( \chi \) Bubble coalescence rate
\( \Omega \) Bubble break-up rate
\( \xi \) Size ratio between an eddy and a particle
\( \varphi \) Volumetric concentration of nanoparticles in nanofluid

**Subscripts**

\( bm \) Brownian motion
\( g \) Gas phase
\begin{align*}
i, j, k & \quad \text{Bubble group number} \\
l & \quad \text{Liquid phase} \\
\text{lm} & \quad \text{Liquid microlayer} \\
nf & \quad \text{nanofluid} \\
v & \quad \text{Vapor phase}
\end{align*}
Chapter 1

Introduction

1.1 Background and Motivation

Nanofluids are engineered colloidal dispersions of nano-scale particles (nanoparticles hereafter) in base fluids. Typical particle materials include oxides (Al$_2$O$_3$, CuO, TiO$_2$, Fe$_2$O$_3$, ZrO$_2$ and SiO$_2$, etc.), electrochemically noble metals (Cu and Ag, etc.) and some other compounds (SiC, etc.). The base fluids usually include water, ethylene glycol, propylene glycol, engine oil, etc. In recent years, the rapidly advanced nanotechnology has spawned into many new engineering applications by implementing nanofluids, such as nuclear reactors (Buongiorno and Hu, 2009), ultrafast cooling systems (Jha et al., 2015), solar collectors (Mahian et al., 2013), microelectronics (Zhang et al., 2013) and automotive industries (Peyghambarzadeh et al., 2013). Nanofluids have been treated as perfect substitutions for pure liquids as energy transfer media, due to their merits in heat transfer capabilities, such as thermo-physical properties, single-phase convective heat transfer, and nucleate boiling heat transfer. Beyond that, the high surface to volume ratio, low mass, and low inertia of nanoparticles enable nanofluids to be highly colloidal stable and less erosional, which can bring synergies of higher mass/energy transfer rate (Abdel-Fattah and El-Genk, 1998).
1.1.1 Thermo-physical Properties

The key thermo-physical property of fluids is thermal conductivity which has received the most attention in the nanofluid research community over the past decade. Using Hamilton-Crosser’s effective thermal conductivity model, Choi and Eastman (1995) firstly investigated the increased thermal conductivity of 20 vol% CuO/water nanofluids. A factor of 3.5 over the base water value was predicted. Later in the experiments conducted by Eastman et al. (1997), the thermal conductivity of 5 vol% CuO/water nanofluids showed an incensement up to 60% compared with water. Following the pioneering work of Choi and his fellows, a number of researchers joined in exploring the anomalous enhancement of the thermal conductivity experimentally and/or theoretically with various combinations of nanoparticles and base liquids (Eastman et al., 2001; Xie et al., 2002; Das et al., 2003; Jang and Choi, 2004).

The basic understanding of the mechanism underlying thermal conductivity enhancement is due to nanoparticles’ high thermal conductivity. However, experimental results indicated that new heat transport mechanisms exist in nanofluids. Murshed et al. (2005) experimentally investigated the thermal conductivity of TiO$_2$/water nanofluids. The thermal conductivity of nanofluids was found to increase remarkably with increasing volume concentration of nanoparticles. Besides the influence of nanoparticle concentration, Chon et al.’s study (2005) showed that nanoparticle size and shape also has significant impact on the thermal conductivity enhancement. In their study, an experimental correlation for the thermal conductivity of Al$_2$O$_3$/water nanofluids was proposed as a function of nanoparticle size ranging from 11 nm to 150 nm over a wide range of temperature, from 21 to 71 °C. The thermal conductivity of nanofluids exhibited strongly temperature- and size-dependent characteristics. In order to explain the spectacular enhancement, various theories have been proposed such as Brownian motion (Jang and Choi, 2004), the formation of an interfacial nano-layer around particles (Yu and Choi, 2003), the percolation-like behaviour (Foygel et al., 2005) and the micro-convection and lattice vibration of nanoparticles (Gupta et al., 2006).

To predict the effective thermal conductivity, some models and/or empirical correlations have been proposed. One of the most popular theoretical models is
developed by Hamilton and Crosser (1962). Their model was a function of the thermal conductivity of both the base fluid and the particle volume fraction of the particles, and the shape of the particles. Since temperature has a significant effect on the thermal conductivity enhancement, recently, Khanafer and Vafai (2011) correlated the experimental data of Al$_2$O$_3$/water nanofluid at various temperatures, nanoparticle size, and volume fraction and proposed their own model, which has been validated in a few experiments.

1.1.2 Single-phase Convective Heat Transfer

Single-phase convective heat transfer plays a significant role in various industry sectors. Nanoparticles have been shown to enhance the convective heat transfer by an increasing number of studies in the past decade. For example, the characteristics of the fully developed convective heat transfer and flow for Cu/water nanofluids through a straight tube with inner diameter of 10 mm was experimentally investigated by Xuan and Li (2003). A constant heat flux condition along the tube wall was imposed using DC heating. Results showed that the nanofluids gave substantial enhancement of heat transfer rate compared to pure water. Enhancement was also found in Wen and Ding’s experiments (2004) where γ-Al$_2$O$_3$ nanoparticles and water flowed through a copper tube in the laminar flow regime. The enhancement was found to be particularly significant in the entrance region suggesting that the enhancement of the thermal conductivity is not the only reason. The non-uniform distribution of thermal conductivity and viscosity filed and the reduced thickness of thermal boundary layer can also influence the convective heat transfer of nanofluids. The hypothesis is then confirmed by Kim et al. (2009) who found the convective heat transfer coefficient for the amorphous carbonic/water nanofluid, under laminar flow, increased by 8% even if its thermal conductivity was similar to that of water. Daungthongsuk and Wongwises (2007) further pointed out the other two plausible reasons for the forced convective heat transfer enhancement of the nanofluids: the increased fluctuations induced by the chaotic movement of nanoparticles and the accelerated energy exchange process due to the extra turbulence. In 2006, Buongiorno developed a two-component four-equation nonhomogeneous equilibrium model for mass, momentum, and heat transport in
nanofluids. They proposed that due to the effects of the temperature gradient and thermophoresis, the viscosity of nanofluid may decrease significantly within the boundary layer. This decrease can lead to the enhancement of convective heat transfer of nanofluids.

Recently, the single-phase convective heat transfer of nanofluids with uniform heat flux or temperature conditions on the wall has been simulated numerically by a number of researchers. For example, Corcione et al. (2012) conducted the simulation under the assumption that nanofluids behave more like single phase fluids than like conventional solid-liquid mixture. Thus all the convective heat transfer correlations available in the literature for single-phase flows were extended to nanoparticle suspensions. In summary, this single phase assumption depends largely on the base fluid, nanoparticle materials, concentration and size. When the particle is extremely small and the volume concentration is very low, the nanofluids can be treated as pure fluids. This hypothesis can be found in a number of other studies (Palm et al., 2006; Demir et al., 2011).

1.1.3 Boiling Heat Transfer

Since You et al. (2003) firstly reported a considerable critical heat flux (CHF) enhancement in Al₂O₃/water nanofluid pool boiling with particle concentration ranging from 0 g/l to 0.05g/l, an increasing number of research groups around the world joined in the investigation of heat transfer characteristics of nanofluids in boiling and published in abundance. Nucleate boiling heat transfer and CHF are the main subjects explored. Significant CHF enhancement has been reported consistently, but the maximum achievable enhancement varies depending on the adopted nanoparticle concentration, nanoparticle material, base liquid and heater size and material. Since the enhanced CHF can afford a higher safety margin, nanofluids have been expected to be ideally suited for practical thermal systems where high heat flux removal is needed, such as nuclear reactors and high-power electronic devices.

However, the nucleate boiling heat transfer is controversial, with some studies reporting no change of heat transfer in the nucleate boiling regime, some reporting heat transfer deterioration, and others heat transfer enhancement. It has been revealed that
one of the influencing factors of the heat transfer coefficient (HTC) of nanofluid boiling is the particle concentration. Kwark et al. (2010) found with increasing nanoparticle concentration, the Al$_2$O$_3$/water nanofluids showed a noticeable degradation in the boiling heat transfer coefficient but have exhibited an enhanced CHF value (up to 80% when nanoparticle concentration reached 0.0007 vol%). Further increase in the concentration produced no further CHF enhancement but degraded the boiling heat transfer. Heris (2011) experimentally investigated the boiling heat transfer of the CuO/ethylene glycol-water (60/40) nanofluid. The results indicated that a considerable boiling heat transfer enhancement has been achieved, specifically that the enhancement had increased with increasing nanoparticles concentration and reached 55% at a nanoparticle concentration of 0.5 vol%. Similar trend of the dependence on nanoparticle concentration of HTC has also been found in Krishna et al.’s (2011) study where Cu/water nanofluids were employed. Their results further showed that the maximum enhancement, when the concentration of Cu nanoparticles increased from 0.01 to 0.1 vol%, was 50% and 20%, respectively on smooth and rough heaters. This indicated that the surface roughness of heaters may be another influencing factor that determines the heat transfer in nanofluids. The temperature of the bulk flow field may influence the HTC as well. This conclusion can be drawn in Taylor and Phelan’s study (2009) where the nucleate boiling heat transfer of Al$_2$O$_3$/water nanofluid was enhanced by 25~40%, but subcooled boiling was deteriorated, compared with the pure-water baseline.

In addition to the above experimental observations, nanofluids exhibit more unique features. It was experimentally observed that during the boiling process of nanofluids, suspending nanoparticles can deposit on the heater surface forming a porous layer by Kim et al. (2006b) who conducted the experiments with several dilute nanofluids (Al$_2$O$_3$/water, ZrO/water and SiO$_2$/water with concentration of 0.01 vol%). Their results also revealed that the porous layer of nanoparticles not only changed the surface roughness (Das et al., 2003) but also had impact on surface wettability (Kim et al., 2007). In addition, near-wall hydrodynamics such as bubble generation, growth and detachment on heater surface were also found deferent in nanofluid boiling (Gerardi et al., 2011). Not only in the near-wall region, the two-phase flow structures in bulk flow
field of have been found to be changed as well. For example, the void fraction in horizontal flow boiling of ZnO/water nanofluids (0.001~0.01 vol%) measured by Rana et al. (2014) showed a decrease up to 86% of that in water. With increasing nanoparticle concentration and flow rate, the void fraction decreases, whereas it increases in heat flux.

For all the addressed features, major knowledge gaps remain in the study of gas-nanofluid bubbly flows. In particular, for nanofluid boiling, while numerous experimental studies of boiling heat transfer have been conducted, numerical studies have not. So far, the underlying mechanisms that how nanoparticles influence the boiling heat transfer have not yet been fully understood. Mathematic models capable of accurately describing the boiling process and effectively predicting the boiling heat transfer in nanofluids are still absent from the open literature, which hinders nanofluid’s further application in industry. Even though a number of models, such as heat flux partitioning (HFP) model, two-fluid model and MUltiple-SIze-Group (MUSIG) model have been previously developed and widely employed in simulating two-phase gas-liquid bubbly flows, without the in-depth study of the mechanism, their applicability to nanofluids is still questioned. Therefore, a numerical study is needed to reveal the role of nanoparticles, and further develop a mathematic model for gas-nanofluid bubbly flows.

1.2 Objectives

The primary goal of this study is to develop a numerical model which is capable of giving a full description and an accurate prediction of the boiling flows of nanofluids. In order to achieve this goal, the following sequential activities have been conducted:

- Review experimental findings in the literature to explore the characteristics of boiling flows of dilute nanofluids and collect data of HTC with various experimental conditions for heat flux, and type of nanofluids (materials and concentrations).
- Examine the feasibility of the existing models such as heat flux partitioning (HFP) model, the two-fluid model, MUltiple-SIze-Group (MUSIG) model in
effective modelling of gas-nanofluid flows by comparing the experimental data with numerical results.

- Analyse the influencing factors in the CHF enhancement and HTC alteration in nanofluid boiling flows and quantify their influences through numerical method.
- Develop new correlations or models for gas-nanofluid flows with or without heat transfer.

1.3 Thesis Outline

The aim of this chapter is to provide a brief description of the research work, started with the background and motivation of the research in nanofluids. Then the objectives are described and explained subsequently. An outline of the thesis based on each chapter is included at the end of this chapter.

Chapter 2 firstly introduced the fundamentals of boiling. The characteristics of heated surface, bubble dynamics in the near-wall region and two-phase flow structures should be the main focuses for the study in pool and flow boiling, respectively. Experimental studies of the unique features observed in gas-nanofluid bubbly flows are then reviewed. The review begins with experimental findings of the boiling heat transfer such as critical heat flux (CHF) and heat transfer coefficient (HTC). Potential influencing factors are then analysed, including the thermo-physical properties, characteristics of the heated surface, near surface hydrodynamics and bulk flow field hydrodynamics. In the last section of this chapter, the preliminary mathematic models, including the heat flux partitioning (HFP) model, the two-fluid model, and the MUSIG model are introduced.

Chapter 3 covers the HFP modelling and analyses of the heat transfer in pool boiling of dilute nanofluids. A study of the effects of nanoparticle deposition on boiling parameters such as nucleation site density, bubble departure diameter and bubble departure frequency are conducted. New correlations of these boiling parameters are proposed. In addition to that, after analysing the process of nanoparticle deposition in micro-scale, a new heat flux partitioning (HFP) model considering the heat transfer by nanoparticle Brownian motion in the microlayer is also developed. Comparison of numerical results against experimental data shows a good consistency.
Chapter 4 provides numerical approaches to investigate the two-phase flow structures of isothermal gas-nanofluid bubbly flows with the two-fluid model and MUSIG model, respectively. It is suggested that in a bubbly flow system, the existence of interfaces allows the spontaneous formation of a thin layer of nanoparticle assembly at the interfaces, which significantly changes the interfacial behaviours of the air bubbles and the roles of the interfacial forces. Thus, one of the most important tasks when modelling bubbly flows of gas-nanofluid using the two-fluid model is to reformulate the interfacial transfer terms according to the interfacial behaviour modifications induced by nanoparticles. Since assembled nanoparticles also have effects on bubble coalescence process, it is also pointed out that modelling the coalescence process in nanofluids is essential to the successful simulation of gas-nanofluid bubbly flows using MUSIG model.

Chapter 5 focuses on mechanistic study of bubbly hydrodynamics in gas-nanofluid bubbly flows. In particular, the underlying mechanism that how nanoparticles affect the interfacial forces acting on bubbles such as the drag force and lift force and what the role that nanoparticles have played in influencing bubble-bubble interaction and further modifying the two-phase flow structures are discussed. Results show that the adsorbed nanoparticles make a bubble behave somewhere between a clean bubble and a solid particle. As a result, flow separation occurs and a slanted wake region forms behind the nanoparticle-adsorbed bubble at a small Reynolds number. Both pressure and viscous stress on the bubble interface become asymmetrically distributed due to the nanoparticle surface concentration. In addition, the interactions between nanoparticles such as electrostatic double layer force and steric repulsion force can not only resist the approach of two bubbles, but also hinder the fluctuation of the liquid film.

Chapter 6 presents the conclusion of this thesis by summarizing the outcomes from chapter 3 to chapter 5 and discusses further investigations required.
Chapter 2

Literature Review

2.1 Overview of Boiling Heat Transfer

Heat transfer process in gas-liquid two-phase flows is accompanied by the presence of a moving and deforming phase interface. Specifically, during boiling process vapour bubbles rapidly form at the solid-liquid interface, detach from the surface when they reach a certain size, and attempt to rise to the free surface of the liquid. According to the bulk fluid motion, boiling is classified as pool boiling, which is under quiescent fluid conditions, or flow boiling, which is under forced-flow conditions.

2.1.1 Pool Boiling

Pool boiling refers to boiling along a heated surface submerged in a large volume of quiescent liquid (Naterer, 2002). As shown in Figure 2.1, pool boiling arises under two types of conditions: electrical heating and thermal heating. With electrical heating, the heat flux can be calculated based on measurements of the applied current and voltage. Thus the heat flux is an independent variable, whereas temperature is a dependent variable. However, in thermal heating, the surface temperature can be set independently of the heat flux. Figure 2.1 also illustrates that in pool boiling any liquid motion is due to free convection and mixing induced by bubble growth and detachment from the heated surface.
The study of pool boiling was pioneered by Nukiyama (1966) who used electrically heated nichrome and platinum wires immersed in liquids in his experiments. Nukiyama noticed that boiling takes different forms, depending on the value of the wall superheat $\Delta T_{\text{sup}} (=T_W-T_{\text{sat}})$, which is the temperature difference between the heater surface and the saturation temperature of the liquid. Four distinct boiling regimes are identified: natural convection boiling, nucleate boiling, transition boiling, and film boiling. These regimes are illustrated on Nukiyama’s boiling curve in Figure 2.2, which is a plot of boiling of heat flux $q$ versus the wall superheat $\Delta T_{\text{sup}}$.

Figure 2.1: Electrical and thermal heating (Naterer, 2002)

Figure 2.2: Nukiyama’s boiling curve (Nukiyama, 1966).
- **Natural convection** (up to A): Free single-phase natural convection occurs from the heated surface to the saturation liquid without formation of bubbles.
- **Nucleate boiling** (A-C): Bubbles nucleate, grow and depart from the heated surface, and further coalesce, mix, and ascend as merged jets or columns of vapour, as wall superheat increases.
- **Transition boiling** (C-D): An unstable (partial) vapour film forms on the heating surface, and conditions oscillate between nucleate and film boiling.
- **Film boiling** (beyond D): A stable layer of vapour forms between the heated surface and the liquid, and blocks the liquid from contacting the surface.

Among these four boiling regimes, nucleate boiling is the most desirable one in practice because high heat transfer rates can be achieved in this regime with relatively small values of $\Delta T_{sup}$, typically under 30 °C for water. During nucleate boiling, vapour bubbles start forming at cavities along the heated surface where a gas or vapour phase already exists. The liquid in microlayer, which is a thin layer underneath the bubble, extract heat from the surface and evaporate. Due to the continuous heating and liquid evaporation, the vapour bubbles keep growing and expanding until the buoyancy force is large enough to lift the bubbles from the cavities. During this process, bubbles ascend and carry away the latent heat of evaporation, while liquid between the bubbles continues to absorb heat by natural convection from the surface (Figure 2.3).

![Figure 2.3: Bubble grow and departure on an active site (Li et al., 2014a).](image)

At large values of $\Delta T_{sup}$, the rate of evaporation at the heater surface reaches such high values that bubbles grow rapidly and eventually merge together. Consequently, a
large fraction of the heated surface will be covered by bubbles, making it difficult for the liquid to reach the heated surface and wet it. Thus, the heat flux increases at a lower rate with increasing $\Delta T_{sup}$, and reaches a maximum at point $C$ in Figure 2.2. The heat flux at this point is the critical heat flux ($q_{max}$, CHF). Nukiyama (1966) noticed that when the power applied to the nichrome wire immersed in water exceeded $q_{max}$ even slightly, the wire temperature jumped suddenly to the melting point of the wire (1500 K) and burnout occurred beyond his control. Therefore, point $C$ on the boiling curve is also called the burnout point. In the design of boiling heat transfer equipment, it is extremely important for the designer to have a good knowledge of the critical heat flux to avoid the danger of burnout.

2.1.2 Flow Boiling

Flow boiling is the boiling process where the fluid is forced to move in a heated pipe (internal flow boiling) or over a surface (external flow boiling) by external means such as a pump as it undergoes a phase-change process. Since there is no free surface for the vapour to escape during internal flow boiling (two-phase flow), the consequent mixing of the liquid and vapour phase make it more complicated in nature and strongly influence the boiling heat transfer. Therefore, flow boiling heat transfer is closely related to the two-phase flow structure of the evaporating fluid. And it exhibits characteristics of both convection and pool boiling. Commonly observed flow structures are defined as two-phase flow patterns. The flow patterns encountered in co-current upflow of gas and liquid in a vertical tube are shown in Figure 2.4.

- **Bubbly flow**: small discrete bubbles in the continuous liquid phase with various shapes and sizes.
- **Slug flow**: with increasing the gas fraction, larger bubbles formed due to collision and coalescence.
- **Churn flow**: with increasing the velocity, the flow becomes unstable and the liquid travels up and down in an oscillatory fashion.
- **Annular flow**: a thin film of liquid on the wall with the gas as the continuous phase in the centre of the tube.
The different stages encountered in flow boiling in a heated tube are illustrated in Figure 2.5 together with the variation of the heat transfer coefficient along the tube. Initially, the liquid is subcooled and forced convection dominates the heat transfer to the liquid. Then the bubbles’ formation and detachment from the heated surface of the tube, and the sequent draft into the mainstream gives the fluid flow a bubbly appearance. With the fluid heated further, the size of the bubbles increase gradually and eventually approach the pipe diameter due to bubble coalescence. The slug of vapour occupy up to half of the volume in the tube until the liquid mainly flows as a film along the walls and the core of the flow consists of vapour only. This is the annular-flow regime, and very high heat transfer coefficients are realized in this regime.

In pool boiling, the vapour flow is largely buoyancy driven. In contrast, forced flow boiling involves bulk motion of the liquid and buoyancy effects. Thus the heat transfer coefficient is less dependent on heat flux than in pool boiling, while its dependence on the local vapour quality appears as a new and important parameter. Both the nucleate and convective heat transfer mechanisms must be taken into account to predict heat transfer data in the flow boiling regime. The local flow parameters such as void fraction, bubble velocity, bubble size and interfacial area concentration become critical to the prediction of heat transfer in flow boiling.
Figure 2.5: Two-phase flow regimes in vertical pipe flow (Naterer, 2002).

How to improve the critical heat flux and the heat transfer coefficient has always been a hot topic in the research of boiling heat transfer. For pool boiling, since the fluid in bulk flow field is almost stationary, the focus is on the heated surface where evaporation and convection mostly occur. Techniques such as sintering, brazing, and flame spraying, which can modify the characteristics of the heated surface have been developed rapidly and numerously to build porous structures on the heated surface and enhance nucleation (Pais and Webb, 1991). Bubble coalescence and interactions between the vapour columns can also affect total heat transfer by changing the convective flow of liquid returning to the heating surface. For flow boiling, as previously mentioned, the heat transfer is closely related to the two-phase flow structure of the evaporating fluid. As the use of nanofluids instead of pure liquids can significantly enhance the boiling heat transfer, a detailed and systematic literature review of experimental findings of gas-nanofluid bubbly flows is needed, in order to develop a comprehensive model.

2.2 Experimental Findings of Nucleate Boiling of Nanofluids

2.2.1 Pool Boiling Heat Transfer

The research in the boiling heat transfer of nanofluid dates back to the experimental study conducted by Yang and Maa (1984). Even though the concept of nanofluid has not been proposed at that time, Yang and Maa discovered an enhancement up to 400%
in HTC for pool boiling of water containing suspended alumina nanoparticles of 50, 300 and 1000 nm in size with concentrations of 0.03 and 0.14 vol% on a horizontal 3.2 mm diameter cylindrical heater. However, for a pool boiling of Al₂O₃/water nanofluids with various nanoparticle concentrations (0.1~4 vol%) on a 20 diameter steel heater, the experimental results of Das et al. (2003a) showed a higher value of wall superheat ΔT_{sup} at a given heat flux which indicated that HTC of the base fluid (water) has been deteriorated with the addition of nanoparticles (Figure 2.6). It has been further observed with increasing particle concentration, the degradation in boiling performance takes place which increases the heater surface temperature. This means that without changing the boiling temperature the nanofluid can cause harm to cooled surface if boiling limit is reached.

![Figure 2.6: Boiling curve of nanofluids on: (a) smooth heater surface (R_a=0.4μm); (b) roughened heater surface (R_a=1.15μm) (Das et al., 2003a).](image)

Interestingly, almost at the same time in 2003, a considerable CHF enhancement (nearly 200%) in Al₂O₃/water nanofluids with nanoparticle concentration ranging from 0.001g/l to 0.05g/l was firstly observed by You et al. (2003). The obtained boiling curves of the pure water and nanofluids are illustrated in Figure 2.7. As shown in the figure, adding extremely small amount of nanoparticles (0.001g/l) in the pure water illustrated a sizable increase in q_{max} value, from 540 to 670kW/m². When the concentration is greater than 0.005g/l, CHF was increased consistently by about 200%
compared to that of the pure water case. Despite the huge CHF enhancement, the boiling heat transfer coefficient values of all concentrations including pure water appeared to be the same.

Figure 2. 7: Boiling curve of pure water and Al$_2$O$_3$/water nanofluids (0.001g/l to 0.05g/l) (You et al., 2003).

Following the pioneering work of Yang and Maa (1984), Das et al. (2003) and You et al. (2003), more than 200 papers focusing on nanofluid boiling have been published in open literature. Most of these studies reported the increase of CHF up to 200% in nanofluids. However, there has been considerable disagreement over the value of the boiling heat transfer coefficient (HTC) of gas-nanofluid flows. Nearly even three-way split in experimental results have been found: enhancement, deterioration, and little or both enhancement and deterioration.

2.2.1.1 Enhancement

Tu et al. (2004) tested Al$_2$O$_3$ nanofluids on a ‘nanoscopically smooth’ vapor-deposited heating surface. Results showed HTC enhancement (~64%) and a fourfold increase in nucleation sites. Similar HTC enhancement was also found in Wen and Ding’s experiment (2005) where the pool boiling of Al$_2$O$_3$/water nanofluids on a stainless steel disc inside a cylindrical vessel was investigated. The pool boiling HTC significantly enhanced with the increasing particle concentration in nanofluids compared to water,
resulting in 40% enhancement for 1.25 wt%. Later in 2007, Liu et al. (2007) tested CuO/water nanofluids on smooth micro-grooved surfaces at various pressures and nanoparticle concentrations. They found significant enhancements (~25% at 100kPa and 150% at 7.4kPa) until the mass concentration exceeded 1%—after which enhancement decreased. Truong et al. (2007) also found very high enhancements (up to 68%) in heat transfer during pool boiling experiments with SiO₂ and Al₂O₃/water nanofluids. The largest enhancement of HTC was observed in the experimental study of Wen et al (2011), where the pool boiling of 0.001 vol% Al₂O₃/water nanofluids on smooth heater surfaces exhibited a two-fold increase in boiling heat transfer coefficient under low heat flux conditions (Figure 2.8).

![Figure 2.8: Comparative boiling experiments on the smooth surface (Wen et al., 2011).](image)

In summary, the studies (Tu et al., 2004; Wen and Ding, 2005; Liu et al., 2007; Kathiravan et al., 2009; Soltani et al., 2009; Liu et al., 2010; Wen et al., 2011; Yang and Liu, 2011; Kole and Dey, 2012; Mourguès et al., 2013; Raveshi et al., 2013) of dilute nanofluids showed enhancement ranging from 15% to 200% in nucleate boiling heat transfer. A wide variety of materials and geometries for nanoparticles and heaters were used.

### 2.2.1.2 Deterioration

Bang and Chang (2005) investigated the pool boiling characteristics of Al₂O₃/water nanofluids (0.5~4 vol%) on horizontal and vertical smooth heaters (Rₑ=37nm). Their results showed 25-50% deterioration in HTC with the increase in nanoparticle concentration. Milanova et al. (2006) tested several types of nanofluids: Al₂O₃, SiO₂,
and CeO$_2$ at 0.5 vol%. With changing PH in pool boiling experiments, the authors observed a decrease in nucleate boiling heat transfer. They also noted that their nichrome (NiCr) wires were oxidized and that there was significant particle deposition during the boiling experiments. Jackson et al. (2006) tested Au nanofluids (0.003 vol%) on a Cu block at various pressures. Overall, Jackson et al. found that the HTC was reduced 25% while the CHF increased 2.5 times. Their results further revealed that the surface roughness was increased by the nanofluids. In 2007, Kim et al. (2007) also tested several nanofluids (Al$_2$O$_3$, ZrO$_2$, SiO$_2$, 0.001~0.1 vol%) on stainless steel wires and plates. Even though due to the lack of exact data of the temperature curve for stainless steel, degradation of the HTC was found. A similar phenomenon that a significant amount of particles was deposited on the heated surface was also observed by the authors using scanning electron microscope (SEM) analysis. They attributed the HTC degradation in nanofluids to the deposited nanoparticles. A deterioration in the HTC as a result of particle deposition was also discovered by Kwark et al. (2010) in their pool boiling experiments with Al$_2$O$_3$/water nanofluids on a horizontal copper block. They found that the HTC deteriorated with coating the boiling surface. For this group of papers (Das et al., 2003a; Bang and Chang, 2005; Jackson, Borgmeyer et al., 2006; Kim et al., 2007; Park et al., 2009; Trisaksri and Wongwises, 2009; Kathiravan et al., 2010; Kwark et al., 2010; Phan et al., 2010; Jung et al., 2012; Sheikhhbahai et al., 2012; Shahmoradi et al., 2013; Mori et al., 2015), deterioration of 0–50% have been found.

### 2.2.1.3 Both Enhancement/Deterioration

As previously mentioned, in a few papers both increased and decreased heat transfer during the tests has been found. Witharana (2003) studied the heat transfer in Au/water nanofluids (0.001 wt%) and SiO$_2$/water-EG nanofluids boiled in a cylindrical vessel under atmospheric pressure, respectively. 21% enhancement of the boiling HTC in Au/water nanofluid was reported, while the SiO$_2$/water-EG nanofluids showed a HTC decrease compared to the base fluids. Narayan et al. (2007) tested Al$_2$O$_3$/water nanofluids on vertical tubular heaters of various surface roughness (48, 98 and 524 nm). It has been observed that with the rough heater ($R_a$=524 nm), heat transfer is
significantly enhanced (~70% at 0.5 wt%). With the smooth heater ($R_a=48$ nm), heat transfer is significantly deteriorated (~45% at 2 wt%). In order to have an insight into the impact of surface roughness on boiling heat transfer, a ‘surface interaction parameter (SIP)’ was defined, which was simply the surface roughness ($R_a$) divided by the average particle diameter. Not only the surface roughness, the nanoparticle concentration also plays an important role in the heat transfer of nanofluid boiling. Chopkar et al. (2008) investigated the nucleate pool boiling of ZrO2/water nanofluid on a Cu plate in a borosilicate tube. Results found the HTC increased at low nanoparticle concentrations, whereas when the concentration increased, it was observed decreasing until becoming lower than that of pure water. The boiling time was also found significant to the total heat transfer rate in nanofluids. Okawa et al. (2012) investigated the boiling time effects with TiO2/water nanofluids (0.000094~0.047 vol%) on a copper block. The experimental results showed that the heat transfer first decreased, then increased, and finally reached an equilibrium situation. Besides the influence of boiling time, the pressure may be another determined factor. Naphon and Thongjing (2014) investigated the influence of TiO2 nanoparticles on the boiling heat transfer of refrigerant R141-b and ethyl alcohol with a brass cylindrical heater. At high heat flux, the boiling HTC was deteriorated with the addition of nanoparticles. However, under high boiling pressure, the HTC increased.

This group of papers shows mixed or discrepant incongruent experimental results about the characteristics of nanofluid boiling heat transfer. According to the literature, the value of HTC is influenced by the nanoparticle concentration (Chopkar et al., 2008; Shoghl and bahrami, 2013), material (Witharana, 2003) and size (Xu and Zhao, 2014) in combination with the heater surface characteristics such as the surface roughness (Narayan et al., 2007; Harish et al., 2011; Wen et al., 2011), and some external factor including the flow pressures (Liu et al., 2007; Liu et al., 2010; Naphon and Thongjing, 2014) and boiling duration (Stutz et al., 2011; Okawa et al., 2012).

2.2.2 Influencing Factors

Vafaei and Borca-Tasciuc (2013) summarized that theoretically the boiling heat transfer depends on factors related to the liquid and solid surface properties including:
(a) physical properties of the liquid such as surface tensions, viscosity, thermal conductivity, specific heat, liquid and vapour densities, vapour and liquid enthalpies; (b) characteristics of heated substrate such as roughness, homogeneity, structure, surface chemistry, which affect the active nucleation site density, equilibrium, receding, and advancing contact angles, and (c) near surface hydrodynamics such as departure bubble volume, bubble frequency, and hot/dry spot dynamics.

2.2.2.1 Thermo-physical Properties

To evaluate the roles of the thermophysical properties of nanofluids in boiling performance, two major properties were examined: the surface tension $\sigma$ and the thermal conductivity $k$. The suspended nanoparticles in a base liquid decrease the surface tension of the fluid significantly. Since the surface forces acting on the bubble, including buoyancy, weight and surface tension at the nucleation site, are responsible for the bubble’s departure, such a reduction in surface tension decreases the radius of bubble, and therefore, more active nucleation sites on the heating surface occur, which enhances the boiling HTC (Kim and Kim, 2009; Yang and Liu, 2011; Raveshi et al., 2013). Das et al. (2003a) measured the thermal conductivity with particle concentration and temperature using the temperature oscillation technique. A substantial increase in thermal conductivity of nanofluids is observed, as shown in Figure 2.9. With such a substantial increase (~60% at saturation temperature), Das et al. (2003a) pointed out that nanofluids are expected to enhance heat transfer during boiling, considering fluid conduction in microlayer evaporation under the bubble as well as in reformation of thermal boundary layer at the nucleation site plays a major role in heat transfer during boiling.

Indeed, the conduction heat transfer is very important at the thin fluid layer on the heating surface and, an increase in the thermal conductivity is one of the reasons for the boiling HTC enhancement observed in other researches, as well as an increase in the stability of nanofluid suspensions (Soltani et al., 2009). However, the thermal conductivity is very dependent on the nanoparticle concentration. For dilute nanofluids with low nanoparticle loadings (<0.1 vol%), the measured thermal conductivity of nanofluid was found to be the same as that of water (Williams et al., 2008).
Comparisons of thermo-physical properties between water and Al₂O₃ and SiO₂/water dilute nanofluids (<0.1 vol%) are given in Table 2.1.

Table 2.1 Comparison of thermo-physical properties between water and dilute nanofluids (Kim, 2009)

<table>
<thead>
<tr>
<th>Fluids</th>
<th>Thermal Conductivity (W/m. K) at 378 K</th>
<th>Kinematic Viscosity (mm²/s) at 413 K</th>
<th>Surface Tension (mN/m) at 378 K</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water</td>
<td>0.60 ± 0.0067</td>
<td>0.6577 ± 0.0046</td>
<td>67.7 ± 1.2</td>
</tr>
<tr>
<td>0.001 vol% Alumina</td>
<td>0.61 ± 0.0067</td>
<td>0.6640 ± 0.0003</td>
<td>67.0 ± 0.2</td>
</tr>
<tr>
<td>0.01 vol% Alumina</td>
<td>0.62 ± 0.0110</td>
<td>0.6681 ± 0.0013</td>
<td>47.6 ± 1.1</td>
</tr>
<tr>
<td>0.1 vol% Alumina</td>
<td>0.58 ± 0.0133</td>
<td>0.6894 ± 0.0003</td>
<td>40.9 ± 0.3</td>
</tr>
<tr>
<td>Water</td>
<td>0.60 ± 0.0067</td>
<td>0.8900</td>
<td>67.7 ± 1.2</td>
</tr>
<tr>
<td>0.001 vol% Silica</td>
<td>0.61 ± 0.0058</td>
<td>0.8846 ± 0.0027</td>
<td>72.1 ± 0.08</td>
</tr>
<tr>
<td>0.01 vol% Silica</td>
<td>0.62 ± 0.0033</td>
<td>0.8857 ± 0.0049</td>
<td>72.4 ± 0.06</td>
</tr>
<tr>
<td>0.1 vol% Silica</td>
<td>0.58 ± 0.0100</td>
<td>0.8929 ± 0.0011</td>
<td>72.2 ± 0.07</td>
</tr>
</tbody>
</table>

Figure 2.9: Thermal conductivity enhancement of nanofluids as a function of temperature (Das et al., 2003a).

Even though the thermal-physical properties differ negligibly from those of pure water, as shown in Figure 2.8, the enhancement of the boiling HTC of 0.001 vol% Al₂O₃/water nanofluids is as much as 200% from pure water. This indicates that the change in boiling characteristics of nanofluids cannot be explained in terms of property change alone.
2.2.2.2 Characteristics of the Heated Surface

Surface roughness

In the experiments conducted by Das et al. (2003a), a considerable reduction in the surface roughness takes place after boiling nanofluid (Figure 2.10). They attributed this roughness change to nanoparticles sitting on the relatively uneven heater surface. According to Das et al. (2003a), since the sizes of nanoparticles (20~50 nm) are one to two orders of magnitude smaller than the roughness (0.2~1.2 μm) of the heating surface, the trapped nanoparticles change the surface characteristics making it smoother. In contrast to Das et al.’s analysis, experimental observations in Bang and Chang’s study (2005) exhibited an increase of surface roughness in the boiling of Al₂O₃/water nanofluids (0.5 and 4 vol%). The surface roughness of the heater surface increased with the increasing nanoparticle concentration, as shown in Figure 2.11.

![Figure 2.10: Surface roughness of the smoother heater surface: (a) before boiling; (b) after boiling with nanofluids (Das et al., 2003a).](image)

After analysing their experimental results as well as Das et al.’s findings, Bang and Chang (2005) proposed that the decrease or increase depends on both original surface condition and the size of nanoparticles. If the original surface roughness is smaller than nanoparticles, it can be increased as Bang and Chang’s results. Reversely, if the original surface roughness is larger than nanoparticles, it can be decreased as Das et al.’s results. Both Das et al. (2003a) and Bang and Chang (2005) believed that the nanoparticle’s attachment, which can be considered as a kind of fouling, to the heated surface is the main cause of the roughness change, and consequently the altered heat
transfer coefficient. However, either of them did not provide any direct and clear evidence of deposited nanoparticles on the heater surface, or explanation how deposited nanoparticles influence the boiling heat transfer.

Figure 2.11: Surface roughness of: (a) clear heater \( (R_a=37.2 \text{ nm}) \); (b) heater submerged in 0.5 vol\% alumina nanofluids \( (R_a=67.6 \text{ nm}) \); (c) in 4 vol\% alumina nanofluid \( (R_a=227.7 \text{ nm}) \) (Bang and Chang, 2005).

**Surface wettability**

The microstructure and topography of the heated surface modified by the deposition of suspended nanoparticles during the boiling of nanofluids was firstly published by Kim et al. (2006a), as shown in Figure 2.12. They found that there is almost no difference between the upside and downside of the heating wire in the test pool in terms of the deposition of nanoparticles. This means that the formation of nanoparticle surface coating is mainly attributed to the nucleation of vapour bubbles on the cylindrical wire, not to the gravitational sedimentation of nanoparticles. The deposition of nanoparticles on the heater surface during the boiling process was also observed by Kim et al.
The irregular porous structures formed by deposited nanoparticles were shown in Figure 2.13.

Figure 2.12: Nanoparticle-coated heaters generated by pool boiling experiments of 0.01 vol% nanofluids: (a) TiO$_2$ nanoparticle-coated NiCr wire; (b) Al$_2$O$_3$ nanoparticle-coated NiCr wire; (c) TiO$_2$ nanoparticle-coated Ti wire (Kim et al., 2006a).

Figure 2.13: Scanning electron microscope images of stainless steel surface boiling in: (a) pure water; (b) 0.01 vol% Al$_2$O$_3$ nanofluid; (c) 0.01 vol% ZrO$_2$ nanofluid; and (d) 0.01 vol% SiO$_2$ nanofluid (Kim et al., 2006b).

Figure 2.14: On surface boiled in pure water: (a) pure water droplet; (b) 0.01 vol% Al$_2$O$_3$ nanofluid droplet; on surface boiled in 0.01 vol% Al$_2$O$_3$ nanofluid: (c) pure water droplet; and (d) 0.01 vol% Al$_2$O$_3$ nanofluid droplet (Kim et al., 2006b).

After obtaining the nanoparticle-fouled surface, Kim et al. (2006b) conducted a series of tests of surface properties. An increase in surface roughness was observed. Beyond that, the static contact angle $\theta$ was measured for sessile droplets of pure water and nanofluid to assess the wettability of the fouled heater surface. Figure 2.14 shows that the contact angle decreases from about 70° to about 20° on the fouled surfaces.
Such decrease occurs with pure water as well as nanofluid droplets, thus suggesting that wettability is enhanced by the porous layer on the surface. More details of the static contact angle are given in Table 2.2.

Table 2.2 Static contact angle for water and nanofluids on clean and fouled surfaces (Kim et al., 2007).

<table>
<thead>
<tr>
<th>Fluid</th>
<th>Concentration (vol%)</th>
<th>water</th>
<th>Al2O3 nanofluid</th>
<th>ZrO2 nanofluid</th>
<th>SiO2 nanofluid</th>
</tr>
</thead>
<tbody>
<tr>
<td>Clean surface</td>
<td></td>
<td>79°</td>
<td>80°</td>
<td>73°</td>
<td>71°</td>
</tr>
<tr>
<td>Nanofluid boiled surface</td>
<td></td>
<td>8-36°</td>
<td>14°</td>
<td>23°</td>
<td>40°</td>
</tr>
</tbody>
</table>

**Capillary wicking height**

A photograph of the departure of a large vapour mushroom after bubbles growing on the nanoparticle-coated wire was found to merge at a high heat flux near the CHF in Kim and Kim’s study (2009). As shown in Figure 2.15, although the heater was almost fully covered with growing bubbles, the coated wire effectively prevented departure from the nucleate boiling region. This implies that the supply of liquid on the heater wire is sufficient to endure the high heat flux.

Figure 2.15: Photograph of pool boiling of pure water at 1900 kW/m² (CHF) on a TiO₂ nanoparticle-coated wire with 0.01 vol% nanoparticle concentration (Kim and Kim, 2009).

As analysed by Kim and Kim (2009), this efficient supply of liquid on the heater wire is due to nanoparticle deposition. They also measured the capillary wicking height ($L_c$) of pure water on TiO₂ nanoparticle-coated surface. As shown in Figure 2.16, an $L_c=1.2$ mm was observed at a concentration of 0.001 vol% and then $L_c$ increased steeply to 4.7 mm at 0.01 vol% and to 5.9 mm at 0.1 vol%. This behaviour of $L_c$ demonstrated that the capillary wicking effect was induced by nanoparticle deposition.
Many researchers have tried to explain the deterioration or enhancement of boiling heat transfer as due to the above changes in the characteristics of heated surface during the boiling process. In order to validate this point, some research groups experimentally studied the boiling behaviour of pure water on heated surfaces with nanoparticle coatings. These surfaces are either created during nanofluid pool boiling (Kwark et al., 2010; Ahmed and Hamed, 2012) or pre-fabricated. In Kwark et al.’s experiments (2010), various nanoparticle coatings were generated by submerging a 1cm×1cm heater in saturated Al₂O₃ nanofluid (0.025 g/l). The pool boiling curves for two nanocoated heaters tested in pure water showed that the nanocoatings enhance CHF by 50% and 70%, for the 15 min and 120 min nanocoatings, respectively. In addition, the coating generated over a 120 min period is found to degrade HTC while the coating generated over a shorter 15 min period is found to have minimal effect on HTC. Similar results can be found in Ahmed and Hamed’s study (2012), where pool boiling experiments of pure water on Al₂O₃ nanoparticle-coated surface have been conducted. A significant deterioration in the HTC compared with that of pure water on clean surface was noted (Figure 2.17). All of the above experimental measurements demonstrate the significant impact of nanoparticle deposition on boiling heat transfer.
In order to have an insight into the relationship of boiling heat transfer and structures or properties of nanoparticle-coated surfaces, numerous experiments have been conducted. Narayan et al. (2007) collected several groups of experimental data of heat transfer enhancement in nanofluids and put them in Figure 2.18 versus their self-defined ‘surface interaction parameter (SIP)’ which is the surface roughness ($R_a$) divided by the average particle diameter. A maximum deterioration of 20% in the HTC was experienced at SIP of 1, whereas the HTC was enhanced by 80% at SIP=11.

Figure 2. 17: Boiling curves of pure water on nanoparticle-deposited surfaces (Ahmed and Hamed, 2012).

Figure 2. 18: Effect of surface roughness and particle size on boiling heat transfer (Narayan et al., 2007).
Figure 2.19: Effects of the surface wettability on the heat transfer coefficient (Phan et al., 2009).

Phan et al. (2009) compares the heat transfer performance of pool boiling on the nanoparticle-coated surfaces. The tendency of the presented curves is relatively good and shows a significant change of the HTC by the surface wettability change. Figure 2.19 highlights this observation and shows that the best HTC is obtained with the surface that has a static contact angle close to either 0° or 90° (Phan et al., 2009).

2.2.2.3 Near Surface Hydrodynamics

As aforementioned, pool boiling includes such aspects as bubble nucleation, growth, and departure from heated surface. The heat from the heated surface is transferred to the liquid during series of bubble behaviours. Thus the change of bubble behaviours can have significant impact on the boiling heat transfer.

*Bubble nucleation*

Kim et al. (2006b) analysed the influences of the decreased contact angle. They found a decrease of the contact angle will tend to decrease the number of active cavities. Plausibly this contributes to the decrease in bubble nucleation in nanofluids with respect to pure water, as shown in Figure 2.20. A similar conclusion of the decreased nucleation sties can be found in Wang and Dhir’s experiments (1993), as shown in
Figure 2.21, where the density of active nucleation sites is plotted for contact angles of 90°, 35° and 18°.

Figure 2.20: Nucleate boiling of pure water (left) and 0.01 vol% Al₂O₃ nanofluid (right) at the same heat flux on an electrically heated 0.25 mm diameter stainless steel wire (Kim et al., 2006b).

Figure 2.21: Active nucleation site density versus heat flux for contact angles from 18° to 90° (Wang and Dhir, 1993).

Narayan et al. (2007) revealed that the number of nucleation sites is also related to the surface roughness and particle size. As aforementioned, they defined a surface interaction parameter (SIP). When SIP is much greater than 1, deposited nanoparticles
will multiply nucleation sites by splitting a single nucleation site into multiple ones. When the ratio is smaller than 1, the nucleation sites also can be increased by creating new cavities due to the nanoparticle deposition. However, when SIP is near unity, the deposited nanoparticles sit in nucleation sites and inhibit nucleation.

**Bubble growth**

![High speed camera images of a boiling bubble and corresponding liquid-vapour phase boundary, temperature, and heat flux distribution at the boiling surface in nanofluids (Jung and Kim, 2014).](image)

Figure 2. 22: High speed camera images of a boiling bubble and corresponding liquid-vapour phase boundary, temperature, and heat flux distribution at the boiling surface in nanofluids (Jung and Kim, 2014).
The bubble growth process on the heater surface in pool boiling of nanofluids has been recorded by Jung and Kim (2014) using high-speed and high resolution infrared cameras. A set of temporally and spatially resolved measurements for the bubble dynamics, liquid-vapour phase transitions, temperature and the heat flux were obtained, as shown in Figure 2.22.

Figure 2. 23: Time evolution of the microlayer geometry beneath a growing bubble (Jung and Kim, 2014).

Figure 2. 24: Bubble geometries including the microlayer and dry spot during the bubble growth period (Jung and Kim, 2014).

During the growing period, the bubble grew rapidly and a microlayer formed beneath the bubble; it was gradually depleted from the centre, creating a dry spot. After the microlayer was completely depleted, the equivalent bubble radius was almost unchanged. However, the triple-contact line started to recede toward the centre and the
bubble shape changed from spherical to ellipsoidal, being elongated in the vertical direction. The microlayer thickness was measured, as shown in Figure 2.23. The growth history of the boiling bubble was also given in Figure 2.24.

The growth time of a bubble on nanoparticle-coated surfaces (contact angle 22–85°) was measured by Phan et al. (2009) with a high speed camera. As shown in Figure 2.25, the change of the heat flux from 220 to 300 kW/m$^2$ results in 23% mean decrease of the growth time. Moreover, the growth time increases with the rise of the surface wettability.

![Figure 2.25: Evolution of grow time as a function of contact angle (Phan et al., 2009).](image)

**Bubble departure**

When You et al. (2003) investigated the pool boiling of Al$_2$O$_3$/water nanofluids, bubble departure sizes and frequencies were measured using a high-speed video camera at 240 frames per second. For the photographic measurements, a 390-μm-diameter platinum wire heater was immersed in the pure water and nanofluid of 0.025 g/l concentration. Obvious differences of bubbles departing from the wire heater at 300 kW/m$^2$ can be seen from Figure 2.26, where with the addition of nanoparticles the sizes of bubbles increases while the bubble departure frequency decreases significantly compared to those in pure water.
Figure 2. 26: Bubbles departing from the wire heater immersed in: (a) pure water; (b) Al₂O₃/water nanofluid (0.025 g/l) (You et al., 2003).

The bubble departures on nanoparticle-coated surfaces with various wettability were also investigated by Phan et al. (2009). They found the departure size and frequency are closely related to the surface wettability. As shown in Figure 2.27, with the increase of the surface wettability, larger bubbles were observed. Whereas, a decreased bubble departure frequency can be found with decreasing the contact angle (Figure 2.28).

Figure 2. 27: Bubble departure on heater surfaces with various wettability (Phan et al., 2009).

Figure 2. 28: Bubble departure frequency versus contact angle (Phan et al., 2009).
2.2.2.4 Bulk Field Hydrodynamics

As aforementioned, the addition of nanoparticles in nanofluids has comprehensive effects on the bubble growth and detachment from heater surface. Since liquid motion arises from free convection and mixing due to bubble growth and detachment from the heated surface in pool boiling, it is reasonable to extrapolate that those comprehensive actions induced by nanoparticles can impact the flow field and temperature distribution of boiling flows. Recently, this extrapolation has been demonstrated by Dominguez-Ontiveros et al. (2010). In the experiments, they measured the boiling point temperature and full-field velocity in pool boiling of pure water and Al$_2$O$_3$/water nanofluid (0.002 vol%), respectively. Through comparing the velocity profiles obtained by Dynamic Particle Image Velocimetry (DPIV), the fluid velocity distributions were found to be generally less uniform and decreased in magnitude for the nanofluid cases than for those of the pure water case (Figure 2.29). Additionally, corresponding vorticity distribution maps revealed an increase in magnitude and sign change with increasing nanofluid concentration which indicated a possible increase in fluid circulation due to nanoparticles (Dominguez-Ontiveros et al., 2010). This increased fluid circulation can affect the convective flow of liquid returning to the heated surface which follows the bubble departure. Since convection account for a significant proportion of the total heat transfer, besides the above mentioned three influencing factors, the hydrodynamics in bulk flow field also play an important role in the boiling heat transfer.

![Figure 2.29: Average full-field velocity profile for pool boiling of: (a) pure water; (b) Al$_2$O$_3$/water nanofluid (0.002 vol%) (Dominguez-Ontiveros et al., 2010).](image)

The change of hydrodynamics in bulk flow field was also found in flow boiling. Nayak et al. (2011) studied experimentally the transient and stability behaviours of
boiling two-phase natural circulation loop with water and Al\(_2\)O\(_3\)/water nanofluid (1.0 wt%, approx. 0.25 vol%), respectively. They found that the natural circulation flow behaviours of nanofluid were very close to that of water in single-phase conditions. However, the buoyancy induced flow rates in boiling conditions were relatively higher with nanofluid than with water. Recently, Rana et al. (2014) measured the void fraction in horizontal flow boiling of ZnO/water nanofluids (0.001~0.01 vol%). Results showed that void fraction decreases up to 86% with the use of nanofluid in place of water and it decreases with increasing nanoparticle concentration and flow rate, whereas increase in heat flux (Figure 2.30).

![Figure 2.30: Effect of (a) nanoparticle concentration; and (b) heat flux on void fraction (Rana et al., 2014).](image)

In addition, the modifications of hydrodynamics by nanoparticles were also observed in isothermal two-phase flows. Wang and Bao (2009) investigated the transition of two-phase flow regimes in a vertical capillary tube, using nitrogen as the gaseous phase and water-CuO nanofluid (0.5 wt%, approx. 0.08 vol%) and pure water as the liquid phase, respectively. They found that the bubbly-slug flow regime transition occurred at a lower liquid superficial velocity or a higher gas superficial velocity in the nanofluid than in water (Figure 2.31). This indicated that nanofluids could maintain a bubbly flow pattern with a higher void fraction than pure water, which is undoubtedly of great importance to enhancing two-phase heat and mass transfers, thanks to the larger interfacial area created by the higher void fraction in nanofluids.
Figure 2. 31: Comparisons of the flow pattern transitions among nitrogen-nanofluid, nitrogen-water/SDBS mixture and nitrogen-water (Wang and Bao, 2009).

Figure 2. 32: Comparison of the local two-phase flow parameters: (a) void fraction; (b) bubble velocity; (d) IAC; and (d) mean bubble diameter in the bubbly flow regime (Park and Chang, 2011).
Park and Chang (2011) measured the local distributions of air-liquid bubbly flow parameters in a vertical tube using a conductivity double-sensor probe. Both pure water and Al₂O₃/water nanofluid (0.1 vol%) were used as the working liquids. The results showed that when the operational conditions were exactly the same, the air-nanofluid bubbly flow had a more flattened void fraction distribution, lower bubble velocity, higher interfacial area concentration and small bubble size than those in the air-water flow (Figure 2.32).

2.3 Numerical Modelling of Gas-liquid Flows

In principal, when modelling gas-liquid flows, two distinct considerations have to be taken into account: (i) Heat transfer process during boiling on the heated surface and (ii) Two-phase flow and bubble behaviours in the bulk flow. For category (i), the heat transfer rate during boiling process can normally be calculated by appropriately partitioning the wall heat flux. The heat flux partitioning (HFP) model (Kurul and Podowski, 1990) is thus introduced. For category (ii), it has been demonstrated that the use of two-fluid model (Ishii, 1975) can appropriately predict the local distribution of flow parameters such as void fraction, bubble velocity, bubble size and interfacial area concentration.

2.3.1 Numerical Modelling of Boiling Heat Transfer

A number of mechanistic models have been developed for the prediction of wall heat flux and partitioning. Del Valle and Kenning (1985) concentrated on the formulation of a mechanistic model for nucleate flow boiling by taking into consideration the bubbly dynamics at the heated wall. This model employed some of the concepts developed by Graham and Hentricks (1967) for wall heat flux partitioning during pool nucleate boiling. The mechanistic model by Kurul and Podowski (1990), which is known as the heat flux partitioning (HFP) model, is still the most widely employed in the numerical simulation of boiling heat transfer.

2.3.1.1 Heat Flux Components
Based on the heat flux partitioning (HFP) model by Kurul and Podowski (1990), a heated surface is divided into two regions, one is occupied by the liquid and the other one is affected by the growing and departing bubbles. Convection is the only mechanism of heat transfer in the region occupied by the liquid while two heat transfer mechanisms, namely evaporation and quenching, come to play alternately in the bubble-affected region. The time durations for the evaporation and quenching mechanisms in a bubble period are defined as the bubble growth time \( t_g \) and the bubble waiting time \( t_w \), respectively. Thus the HFP model entails the partitioning of the wall heat flux into three heat flux components: (1) Heat transferred by turbulent convection, \( q_c \); (2) Heat transferred by evaporation or vapour generation, \( q_e \); (3) Heat transferred by conduction to the superheated layer next to the wall (nucleate boiling or surface quenching), \( q_q \).

\[
q = q_c + q_e + q_q \tag{2.1}
\]

The heat flux according to the definition of local Stanton number \( St \) for turbulent convection is given as:

\[
q_c = \frac{\pi}{6} d_b \rho \beta \left(T_w - T_i\right) \tag{2.2}
\]

The heat flux due to vapour generation at the wall in the nucleate boiling region can simply be calculated from Bowring (1962):

\[
q_e = \frac{\pi}{6} d_{bw} \rho \beta f N_a h_f \tag{2.3}
\]

The surface quenching heat flux is determined through the relationship:

\[
q_q = \frac{2}{\sqrt{\pi}} f A_c \sqrt{\frac{L_i}{\alpha_0 \rho \beta}} \left(T_w - T_i\right) \tag{2.4}
\]

In the above equations, \( d_{bw} \), \( f \), \( N_a \), \( A_c \) and \( A_q \) are the bubble departure diameter, bubble departure frequency, active nucleation site density, the area fractions of the heater surface subjected to convection and quenching, respectively.

### 2.3.1.2 Boiling Parameters

Numerous empirical correlations have been proposed for the aforementioned boiling parameters such as active nucleation site density, bubble departure diameter, bubble departure frequency and area fractions.
**Active nucleation site density**

In a boiling flow, bubbles occur within small pits and cavities on the heated surface where these nucleation sites are activated when the surface temperature exceeds the saturation temperature. The number of active small pits and cavities per unit area is called the active nucleation site density. In terms of the bubble nucleation mechanism of Bankoff (1958), the availability of cavities on a heater surface for bubble nucleation is strongly affected by the surface microstructures and wettability. Based on this mechanism and a cone cavity assumption, Yang and Kim (1988) correlated the active nucleation site density to the surface microstructures and liquid contact:

$$N_a = N \int_{\beta_0}^{\beta_0/2} f(\beta) \, d\beta \times \int_{r_{\text{min}}}^{r_{\text{max}}} f(r_c) \, dr_c$$

(2.5)

where, $N$ is the total number of possible nucleation sites available on a unit heater surface area, $f(\beta)$ and $f(r_c)$ are the probability density functions for the cone angle and cavity mouth size, respectively. Unfortunately, due to the diversity and inherent complexity of realistic heater surfaces, it’s not easy to formulate a universal active site density correlation based on Equation 2.5.

As a simplification, the active nucleation site density has been widely correlated to the wall superheat and some other parameters such as the liquid contact angle. Among them, the correlations (Equation 2.6~2.9) proposed by Lemmert and Chwala (1977), Wang and Dhir (1993), Basu et al. (2002) and Hibiki and Ishii (2003) are highly regarded in terms of accuracy for nucleate boiling of pure liquids.

$$N_a = (210(T_{\text{sat}} - T_w))^{1.805}$$

(2.6)

$$N_a = 7.81 \times 10^{-29} C_n (1 - \cos \theta) \left( \frac{2\sigma_{\text{sat}}}{\rho_l h_{fg} \Delta T_{\text{sup}}} \right)^{-6.0}$$

(2.7)

$$N_a = \begin{cases} 0.34 \times 10^4 (1 - \cos \theta) \Delta T_{\text{sup}}^{2.0} & \Delta T_{\text{ONB}} < \Delta T_{\text{sup}} < 15K \\ 0.34 (1 - \cos \theta) \Delta T_{\text{sup}}^{5.3} & 15K < \Delta T_{\text{sup}} \end{cases}$$

(2.8)

$$N_a = 4.72 \times 10^3 \left( 1 - \exp \left( -\frac{\theta^2}{4.17} \right) \right) \left( \exp \left( 2.5 \times 10^{-8} f(\rho^*) \frac{\Delta T_{\text{sup}} \rho_x h_{fg}}{2\sigma_{\text{sat}}} \right) - 1 \right)$$

(2.9)

**Bubble departure diameter**
A number of studies examining bubble growth and detachment have resulted in a number of different empirical correlations for bubble departure. Tolubinsky and Kostanchuk (1970) proposed a simple relationship which evaluated the bubble departure diameter as a function of the subcooling temperature as:

$$d_{bw} = \min \left( 0.0006 \exp \left( -\frac{\Delta T_{sub}}{45} \right), 0.00014 \right)$$  \hspace{1cm} (2.10)$$

On the basis of the balance between the buoyancy and surface tension forces at the heater surface, Frize (1935) proposed a correlation which includes the contact angle of the bubble:

$$d_{bw} = 0.0208 \theta \sqrt{\frac{\sigma}{g \left( \rho_l - \rho_g \right)}}$$  \hspace{1cm} (2.11)$$

**Bubble departure frequency**

For the bubble departure frequency, most correlations have been derived from the consideration of the bubble departure diameter. Cole’s correlation (1960) which was derived assuming a balance between buoyancy and drag (drag coefficient constant) for pool nucleate boiling is a popular expression. It is in the form of:

$$f = \sqrt{\frac{4g \left( \rho_l - \rho_g \right)}{3d_{bw} \rho_l}}$$  \hspace{1cm} (2.12)$$

**Area fractions**

The area fractions of the heater surface subjected to quenching $A_q$ is usually given by:

$$A_q = N_s K \frac{\pi d_{bw}^2}{4}$$  \hspace{1cm} (2.13)$$

where the empirical constant $K$ is used to account for the area of the heater surface influenced by the bubble. A value of $K=4$ is often recommended. However, Kenning (1981) have found values ranging between 2 and 5. Judd and Hwang (1976) ascertained that a lower value, $K=1.8$, best fitted their experimental data. Tu and Yeoh (2002) incorporated a Jacob number ($Ja_{sub}$) based on liquid subcooling dependence:
\[ K = 4.8 \exp\left( -\frac{J_{\text{sub}}}{80} \right) \quad (2.14) \]

### 2.3.2 Numerical Modelling of Bulk Flow

The particular difficulties in modelling the bulk flow field of boiling is due to the presence of interfaces between phases and existing discontinuities coupled with them. Among a number of theoretical models, the two-fluid model (Ishii, 1975) where the dispersed bubbles are treated as a continuous phase is regarded as the most advanced one because of the explicit treatment of the interactions between the phases.

#### 2.3.2.1 Two-fluid Model

**Governing equations**

In the two-fluid model, two sets of conservation equations governing the balance of mass, momentum and energy of liquid and gas phases are solved.

The continuity equation of liquid phase

\[ \frac{\partial}{\partial t} \left( \alpha_l \rho_l \right) + \nabla \cdot \left( \alpha_l \rho_l \mathbf{U}_l \right) = I_{ig} \quad (2.15) \]

The continuity equation of gas phase

\[ \frac{\partial}{\partial t} \left( \alpha_g \rho_g \right) + \nabla \cdot \left( \alpha_g \rho_g \mathbf{U}_g \right) = I_{ig} \quad (2.16) \]

The momentum equation of liquid phase

\[ \frac{\partial}{\partial t} \left( \alpha_l \rho_l \mathbf{U}_l \right) + \nabla \cdot \left( \alpha_l \left( \rho_l \mathbf{U}_l \mathbf{U}_l - \mu_l \mathbf{\nabla} \mathbf{U}_l + \left( \mathbf{\nabla} \mathbf{U}_l \right)^T \right) \right) 
= \alpha_l \left( \rho_l g - \nabla P \right) + F_{ig} + \left( I_{ig} \mathbf{U}_g - I_{ig} \mathbf{U}_g \right) \quad (2.17) \]

The momentum equation of gas phase

\[ \frac{\partial}{\partial t} \left( \alpha_g \rho_g \mathbf{U}_g \right) + \nabla \cdot \left( \alpha_g \left( \rho_g \mathbf{U}_g \mathbf{U}_g - \mu_g \mathbf{\nabla} \mathbf{U}_g + \left( \mathbf{\nabla} \mathbf{U}_g \right)^T \right) \right) 
= \alpha_g \left( \rho_g g - \nabla p \right) + F_{ig} + \left( I_{ig} \mathbf{U}_g - I_{ig} \mathbf{U}_g \right) \quad (2.18) \]

The energy equation of liquid phase
\[
\frac{\partial}{\partial t} \left( \alpha_i \rho_i H_i \right) + \nabla \cdot \left( \alpha_i \left( \rho_i \vec{U}_i H_i - \lambda_i \nabla T_i \right) \right) = h_{lg} A_{lg} \left( T_g - T_i \right) + \left( \Gamma_{lg} H_g - \Gamma_{lg} H_i \right)
\]

(2.19)

The energy equation of gas phase

\[
\frac{\partial}{\partial t} \left( \alpha_g \rho_g H_g \right) + \nabla \cdot \left( \alpha_g \left( \rho_g \vec{U}_g H_g - \lambda_g \nabla T_g \right) \right) = h_{lg} A_{lg} \left( T_i - T_g \right) + \left( \Gamma_{lg} H_i - \Gamma_{lg} H_g \right)
\]

(2.20)

**Inter-phase mass transfer**

In subcooled boiling flow, the source term \( \Gamma_{lg} \) in Equation 2.1 represents the mass transfer rate due to condensation in the bulk subcooled liquid. It can be expressed by:

\[
\Gamma_{lg} = \frac{h_{fg} A_{lg} \left( T_{sat} - T_i \right)}{h_{fg}}
\]

(2.21)

where \( T_{sat} \) and \( h_{fg} \) represent the saturation temperature and the latent heat of vaporization, respectively.

**Inter-phase momentum transfer**

The inter-phase momentum transfer is mainly the interfacial force \( \vec{F}_{lg} \) which generally includes the forces due to viscous drag \( \vec{F}_D \), the lateral lift \( \vec{F}_L \), the wall lubrication \( \vec{F}_W \), and the turbulent dispersion \( \vec{F}_{TD} \), which are defined by the following equations:

\[
\vec{F}_{lg} = -\vec{F}_{gl} = \vec{F}_D + \vec{F}_L + \vec{F}_{ID} + \vec{F}_W
\]

(2.22)

The drag force \( \vec{F}_D \) is one of the most important forces encountered in bubbly flows, and it dominantly controls the relative motion of each phase. The inter-phase momentum transfer between gas and liquid due to drag force is given by:

\[
\vec{F}_D = \frac{3}{4} C_D \frac{d_i}{d_g} \alpha_g \rho_i \left| \vec{U}_g - \vec{U}_i \right| \left( \vec{U}_g - \vec{U}_i \right)
\]

(2.23)

The drag coefficient \( C_D \) in Equation 2.23 is empirically correlated by Ishii and Zuber (1979) to the bubble Reynolds number \( Re_b \) and Eötvös number \( Eo \):
\[
C_D = \begin{cases}
\frac{24}{Re_b} & 0 < Re_b \leq 0.2 \\
\frac{24(1 + 0.1Re_b^{0.75})}{Re_b} & 0.2 < Re_b \leq 1000 \\
\frac{2}{3} Eo^{0.5} & 1000 < Re_b 
\end{cases}
\] (2.24)

\(Re_b\) and \(Eo\) are defined by:

\[
Re_b = \frac{\rho_g (\bar{U}_g - \bar{U}_l) d_b}{\mu_l} \quad (2.25)
\]

\[
Eo = \frac{g(\rho_l - \rho_g) d_b^2}{\sigma} \quad (2.26)
\]

The lift force \(\vec{F}_L\) generally acts in the direction normal to the relative motion of fluid and bubbles, and largely controls the transverse motion of bubbles in a vertical flow. It can be described according to Drew and Lahey (1987):

\[
\vec{F}_L = C_L \alpha \rho_l (\bar{U}_g - \bar{U}_l) \times (\nabla \times \bar{U}_l) \quad (2.27)
\]

The empirical Tomiyama correlation (1998) is generally used to calculate the lift coefficient \(C_L\):

\[
C_L = \begin{cases}
\min [0.288, \tanh(0.121Re_b^{*}, f(\text{Eo}^*))] & \text{Eo}^* \leq 4 \\
f(\text{Eo}^*) = 0.00105\text{Eo}^{*3} - 0.0159\text{Eo}^{*2} - 0.0204\text{Eo}^{*}0.474 & 4 < \text{Eo}^* \leq 10 \\
-0.27 & 10 < \text{Eo}^*
\end{cases}
\] (2.28)

where \(\text{Eo}^*\) is the modified Eötvös number based on the maximum bubble horizontal dimension \(d_H\) that can be computed by using the empirical correlation given by Wellek et al. (1966).

\[
\text{Eo}^* = \frac{g(\rho_l - \rho_g) d_H^2}{\sigma} \quad (2.29)
\]

\[
d_H = d_b \left(1 + 0.163\text{Eo}^{0.757}\right)^{0.1} \quad (2.30)
\]

The wall lubrication force \(\vec{F}_W\) tends to push the bubbles away from the wall. It acts normal to the wall and decays with distance. According to Antal et al. (1991), it is usually given by:
\[
\bar{F}_w = -\frac{\alpha_g \rho_i (U_g - U_1)}{d_b} \max \left(0, C_{w1} + C_{w2} \frac{d_b}{y_w}\right) \bar{n}
\]
(2.31)

where the wall lubrication coefficients take value of \(C_{w1} = -0.01\) and \(C_{w2} = 0.05\) as suggested by ANSYS CFX. This means the force only exists in the region less than 5 bubble diameters from the wall.

The turbulent dispersion force \(\bar{F}_{TD}\) emerges due to the result of diffusion caused by turbulence. It can be expressed by:

\[
\bar{F}_{TD} = -C_{TD} \rho_f k_i \nabla \alpha_i
\]
(2.32)

where the turbulent dispersion coefficient usually take value of \(C_{TD} = 0.1\).

**Inter-phase heat transfer**

The inter-phase heat transfer can be computed through the term \(h_{lg} A_{lg} (T_g - T_l)\). \(A_{lg}\) is the interfacial area per unit volume. For flow of spherical bubbles of diameter \(d_b\) in a liquid, the interfacial area per unit volume is expressed by:

\[
A_{lg} = \frac{6 \alpha_g}{d_b}
\]
(2.33)

The inter-phase heat transfer coefficient \(h_{lg}\), which is the amount of heat energy crossing a unit area per unit time per unit temperature difference, is usually expressed in terms of a non-dimensional Nusselt number \(Nu\), bubble diameter \(d_b\) and the liquid thermal conductivity \(\lambda_l\):

\[
Nu = \frac{h_{lg} d_b}{\lambda_l}
\]
(2.34)

For a bubble in a moving incompressible Newtonian fluid, Hughmark (1967) proposed the most well tested empirical correlation to compute the Nusselt number \(Nu\):

\[
Nu = \begin{cases} 
2 + 0.6 Re_b^{0.5} Pr^{0.33} & 0 \leq Re_b < 766.06 \quad 0 \leq Pr < 250 \\
2 + 0.6 Re_b^{0.62} Pr^{0.33} & 766.06 \leq Re_b < 250
\end{cases}
\]
(2.35)

**Turbulence Model**

There is no standard turbulence model tailored for two-phase turbulent flow. In majority of two-phase flow applications, the standard two-equation \(k-\epsilon\) turbulence
model is employed to resolve the turbulent flow associated with the continuous liquid and dispersed gas phases, even though it has been found to predict relatively high gas void fraction close to the wall (Frank et al., 2004). Considering the bubble-induced turbulence, Sato and Sekoguchi (1981) proposed a new turbulent model where the effective viscosity $\mu_l^{eff}$ of the continues phase in Equation 2.36 consists of the laminar $\mu_l^{lam}$, liquid shear-induced turbulent $\mu_l^{Tl}$ and bubble-induced turbulent $\mu_l^{Tb}$ viscosities.

$$\mu_l^{eff} = \mu_l^{lam} + \mu_l^{Tl} + \mu_l^{Tb}$$  \hspace{1cm} (2.36)

The liquid shear-induced turbulent viscosity is given by:

$$\mu_l^{Tl} = \rho_l C_{\mu} \frac{k^2}{\varepsilon_l}$$  \hspace{1cm} (2.37)

and the bubble-induced turbulent viscosity is evaluated according to:

$$\mu_l^{Tb} = \rho_l C_{\mu b} \frac{\alpha_g d_b}{U - \bar{U}_l} \left| \bar{U}_l - \bar{U}_i \right|$$  \hspace{1cm} (2.38)

in which the constants $C_{\mu}$ and $C_{\mu b}$ take on values of 0.09 and 1.2, respectively. Effective viscosity in the gas phase can now be simply evaluated as:

$$\mu_g^{eff} = \mu_i^{eff} \frac{\rho_g}{\rho_i}$$  \hspace{1cm} (2.39)

2.3.2.2 MUltiple-SIze-Group (MUSIG) Model

In the two-fluid model, the gas phase is characterised by a single mean diameter $d_b$. The bubbles are therefore assumed to have the same size and shape throughout the domain. In reality bubbles in the liquid phase have a wide spectrum of bubble sizes and shapes, particularly, when they break up and coalescence. In order to handle dispersed multiphase flows in which the dispersed phase has a large variation in size, the MUltiple-SIze-Group (MUSIG) model was developed by Lo (1996). It provides a framework in which the population balance method together with the break-up and coalescence models can be coupled together. In the model, the bubbles are divided into $N$ size groups and each of these size groups can be treated as a separate phase in a multiphase flow calculation. This multiphase flow therefore has $N$ sets of continuity equations. For the $i$th group bubbles ($i$th=1~$N$), the continuity equation is as the following:
\[
\frac{\partial}{\partial t} \left( \alpha_s \rho_g f_i \right) + \nabla \cdot \left( \alpha_s \rho_g \vec{U} f_i \right) = S_i - f_i \Gamma_{lg} \tag{2.40}
\]

where \( S_i \) is the rate of mass transfer into the size group due to bubble break-up and coalescence. In subcooled boiling bubbly flows, the term \( f_i \Gamma_{lg} \) represents the mass transfer due to condensation redistributed for each of the discrete bubble classes. The gas void fraction along with the scalar fraction \( f_i \) is related to the number density \( n_i \) and the total volume \( v_i \) of the discrete bubble \( i \)th class as:

\[
\alpha_s f_i = n_i v_i \tag{2.41}
\]

The number density \( n_i \) can be calculated by using the population balance method, which will be introduced in the following section.

**Population Balance Method**

Population balance is a well-established method for computing the size distribution of the dispersed phase and accounting for the break-up and coalescence effects. A general form of the population balance equation is:

\[
\frac{\partial n_i}{\partial t} + \nabla \cdot \left( \vec{U} n_i \right) = S_i \tag{2.42}
\]

where \( S_i' \) is a source term describing the bubble number density variations due to bubble break-up and coalescence. The rate of mass transfer \( S_i \) of the \( i \)th group bubbles in Equation 2.26 can be calculated by:

\[
S_i = S_i' \left( \frac{\alpha_s f_i}{n_i} \right) \rho_g \tag{2.43}
\]

\[
S_i' = B_C + B_B - D_C - D_B \tag{2.44}
\]

where \( B_C \) and \( D_C \) are, respectively, the birth and death rates of the number density of the \( i \)th group bubbles due to coalescence; \( B_B \) and \( D_B \) are the birth and death rates due to break-up. They are formulated as:

\[
B_C = \frac{1}{2} \sum_{k=1}^{N} \sum_{j=1}^{i} n_in_j \chi_{ij} \tag{2.45}
\]

\[
D_C = \sum_{j=1}^{N} n_in_j \chi_{ij} \tag{2.46}
\]
\[ B_B = \sum_{k=1}^{i} \Omega(V_j : V_i) n_j \]  
\[ D_B = \Omega n_j \quad \text{and} \quad \Omega_i = \sum_{k=1}^{N} \Omega_{ki} \]  

where \( \Omega(V_j : V_i) \) is the break-up rate of bubbles of volume \( V_j \) into volume of \( V_i \); \( \chi_{ij} \) is the coalescence rate. These two rates are closely related to the interactions of two bubbles, which are detailed in the next section.

**Modelling of Bubble-bubble Interactions**

The coalescence of two bubbles is often assumed to occur in three steps: (1) the bubbles collide trapping a small amount of liquid between them; (2) the bubbles keep in contact while this liquid film drains; (3) when the contact time is sufficient for the liquid film to drain out down to a critical thickness, the film ruptures, resulting in coalescence. The coalescence process is therefore modelled by a collision frequency \( \theta_{ij} \) of two bubbles and a collision efficiency \( \eta_{ij} \):

\[ \chi_{ij} = \theta_{ij} \eta_{ij} \]  

In a turbulent flow, the collisions between bubbles may be caused by a number of mechanisms such as turbulent fluctuation, laminar shear, wake entrainment, and buoyancy. The former three mechanisms are usually taken into account. The collision frequency \( \theta_{ij} \) is therefore written as:

\[ \theta_{ij} = \theta^T_{ij} + \theta^LS_{ij} + \theta^{WE}_{ij} \]  

where \( \theta^T_{ij} \), \( \theta^LS_{ij} \) and \( \theta^{WE}_{ij} \) represent the collision frequency due to turbulence, laminar shear and wake entrainment, respectively. \( \theta^T_{ij} \) is defined by:

\[ \theta^T_{ij} = \frac{\pi}{4} \left( d_i + d_j \right)^2 \left( u_{\eta_i}^2 + u_{\eta_j}^2 \right)^{1/2} \]  

\[ u_{\eta_i} = \sqrt{2} e^{\nu/3} d_i^{1/3}, \quad u_{\eta_j} = \sqrt{2} e^{\nu/3} d_j^{1/3} \]

The frequency of shear-induced collisions \( \theta^LS_{ij} \) is given by:

\[ \theta^LS_{ij} = \frac{32}{3} \left( d_i + d_j \right)^3 \left| \frac{dU_i}{dR} \right| \]
When bubbles enter the wake region of a leading bubble, they will accelerate and may collide with the preceding one, resulting in bubble coalescence. This mechanism is accounted using the model proposed by Wang et al. (2005):

\[ \dot{\theta}_{ij}^{\text{WE}} = K \Theta d_i^2 u_{si} \]  

(2.54)

where \( K \) is a constant (\( K = 15.4 \)), \( u_{si} \) is the slip velocity defined by:

\[ u_{si} = 0.71 \sqrt{g d_i} \]  

(2.55)

The parameter \( \Theta \) is introduced in consideration that only bubbles larger than \( d_{cr}/2 \) have a wake region effect for bubble coalescence.

\[ \Theta = \begin{cases} \frac{(d_i - d_{cr}/2)^6}{(d_i - d_{cr}/2)^6 + (d_{cr}/2)^6} & d_i \geq d_{cr}/2 \\ 0 & d_i < d_{cr}/2 \end{cases} \]  

(2.56)

\[ d_{cr} = 4 \sqrt{\frac{\sigma}{g(\rho_l - \rho_g)}} \]  

(2.57)

According to Coulaloglou, the collision efficiency \( \eta_{ij} \) is determined by the actual contact time \( \tau_{ij} \) and the drainage time \( t_{ij} \), which is the time required for the liquid film to thin down to a critical thickness.

\[ \lambda_{ij} = \exp \left( -\frac{t_{ij}}{\tau_{ij}} \right) \]  

(2.58)

To estimate the bubble contact time \( \tau_{ij} \) in a turbulent system, the correlation developed by Levich et al. is widely used:

\[ \tau_{ij} = \frac{r_{ij}^{2/3}}{\varepsilon^{1/3}} \]  

(2.59)

\[ r_{ij} = \left( \frac{1}{2} \left( \frac{1 + \frac{1}{r_i}}{r_i} \right) \right)^{-1} \]  

(2.60)

The drainage time \( t_{ij} \) is calculated according to Prince and Blanch:

\[ t_{ij} = \left( \frac{r_y^2 \rho_l}{16 \sigma} \right)^{1/2} \ln \frac{h_0}{h_i} \]  

(2.61)

Luo and Svendsen (1996) developed a theoretical model for the break-up of bubbles in turbulent dispersions. In this model, binary break-up of the bubbles is
assumed and the model is based on the theories of isotropic turbulence. The break-up rate of bubbles of volume $V_j$ into volume size of $V_i$ can be obtained as:

$$
\frac{\Omega(v_j; v_i)}{(1-\alpha_g) n_j} = C \left( \frac{\varepsilon}{d_j} \right)^{1/3} \left( 1 + \xi^2 \right) \exp \left( - \frac{12c_f \sigma}{\beta \rho \varepsilon^{2/3} \sigma d_j} \right) d\xi
$$

where $\xi = \lambda/d_j$ is the size ratio between an eddy and a particle in the inertial subrange and consequently $\xi_{\text{min}} = \lambda_{\text{min}}/d_j$; and $C$ and $\beta$ are determined respectively from fundamental consideration of bubbles break-up in turbulent dispersion systems to be 0.923 and 2.0. The variable $c_f$ denotes the increase coefficient of surface area:

$$
c_f = f_{BV}^{2/3} + (1 - f_{BV})^{2/3} - 1
$$

where $f_{BV}$ is the break-up volume fraction which is between 0 and 1. $f_{BV}=0.5$ refers to equal break-up and $f_{BV}=0$ or 1 refers to no break-up.

### 2.3.3 Main Challenges in Modelling Bubbly Systems of Nanofluids

With regards to modelling boiling heat transfer in nanofluids, the main challenge lies in characterizing the surface modifications and the altered bubble behaviours. In particular, most of the correlations (Equation 2.5~2.14) calculating the boiling parameters in the HFP model are formulated empirically or semi-empirically and validated against a restricted range of experimental data of pure liquids. When the structure and properties of heated surface have been changed as detailed in the aforementioned experimental findings of gas-nanofluid flows, the feasibility of these empirical correlations in modelling nanofluid remains questionable. Assessing their performance and applicability to the experimental data of nanofluids is of great significance to the success of nanofluid modelling. In addition, the process of nanoparticle deposition and its effects on the boiling heat transfer have not been fully explored. As demonstrated by Kim et al. (2007) and Kwark et al. (2009), the deposition of nanoparticles is mainly caused by the evaporation of liquid microlayer. The evaporating microlayer underneath the bubble leaves behind nanoparticles concentrating in it and then adhering to the heater surface when the microlayer is completely vaporized. The concentration of nanoparticles in the microlayer would keep increasing from the bulk value up to 100%. The thermal conductivity could be very high, which may increase the heat transfer...
between the wall and the nanofluid. Thus the partitioning of the wall heat flux in boiling flow of nanofluid might entail more heat flux components.

Due to nanoparticle’s small size, nanoparticles in a nanfluid are thought to be mixed with the base liquid at near-molecular level. A dilute nanofluid thus can be treated as a single liquid in spite of the presence of two distinct phases. This has allowed in the literature developing thermal-fluid dynamic models for nanofluids based on the classic Navier-Stoke equations. The numerical works by Palm et al. (2006), Fard et al. (2010) and Demir et al. (2011) demonstrated that a single-phase CFD model in which the liquid-nanoparticle suspension is treated as a single phase has an accuracy comparable to that of a two-phase model in which the liquid phase and the particle phase are treated separately, provided the suspension properties are properly formulated. Therefore, it is reasonable to extrapolate that the two-fluid model is applicable to the gas-nanofluid bubbly flows with and without heat transfer. However, as mentioned by Ishii and Mishima (1984), the closure correlations describing the interfacial transports are the weakest link in a two-fluid model due to the considerable difficulties in terms of experimentation and modelling. Most of these closure equations are empirical or semi-empirical. In order to achieve an effective modelling of gas-nanofluid bubbly flows, these closure correlations have to be carefully reformulated or selected to account for the specific features induced by nanoparticles. Moreover, as shown in Figure 2.32, under exactly the same injecting conditions, most of the measured bubble diameters in nanofluids were between 2mm to 5mm, which were much smaller than those ranging from 3 mm to 10 mm in water. Since bubble coalescence and break-up dominates the bubble sizes in two-phase flows, questions about the role that nanoparticles have played in resisting coalescence or encouraging break-up arise from this interesting phenomenon. Therefore, remodelling the bubble coalescence and break-up is another challenge for modelling gas-nanofluid bubbly flows.
Chapter 3

Numerical Modelling of Boiling Heat Transfer in Dilute Nanofluids

The main findings of this chapter have been included in:


3.1 A Parametric Study of the Heat Flux Partitioning Model for Nucleate Boiling of Nanofluids

Abstract:
The dramatic boiling heat transfer performances of nanofluids have been widely attributed to the nanoparticle deposition during the boiling process. The deposited nanoparticles significantly change the microstructures and properties of the heater surface, and hence alter the characteristics of bubble nucleation and departure. Therefore, it is crucial to take into account the effects of nanoparticle deposition when modeling nucleate boiling of nanofluids using the heat flux partitioning (HFP) model (Kurul and Podowski, 1990). In this study, new closure correlations were incorporated for the nucleate boiling parameters including the active site density, the bubble departure diameter and frequency. Parametric studies were performed through 2-D computations to analyze the effects of surface wettability enhancement, the nanoparticle material and size, respectively. The results demonstrated that through appropriate considering the modifications induced by nanoparticle deposition, the HFP model achieved a satisfactory agreement with the experimental data available in the literature, and provided a more feasible and mechanistic approach than the classic Rohsenow correlation for predicting nucleate pool boiling of nanofluids.

3.1.1 Introduction

As a new type of engineered fluids, nanofluids have gained an increasing attention due to their enhanced properties associated with heat transfer (Choi and Eastman, 1995). Since 2003 when Das et al. (2003a) and You et al. (2003) pioneered the studies on boiling of nanofluids, an exponentially increasing number of analogous investigations have been conducted with the aim to reveal the mechanisms underlying the dramatic heat transfer performances and novel phenomena observed in boiling nanofluids.

With a view to the practical feasibility, dilute nanofluids, typically with a nanoparticle concentration lower than 0.1% by volume, are generally preferred due to their improved colloidal stability and negligibly altered physical properties from those of their pure base liquids. Kim et al. (2007) measured the properties of several dilute
aqueous nanofluids (Al₂O₃/water, ZrO/water and SiO₂/water with concentrations of 0.001 vol%, 0.01 vol% and 0.1 vol%) and compared them with those of pure water. The results demonstrated that the saturation temperature of these nanofluids was within ±1 °C of that of pure water while their surface tension, thermal conductivity and viscosity were negligibly changed. However, significant critical heat flux (CHF) enhancement up to 60% was detected in these nanofluids. These specific features of dilute nanofluids allow the minimum modification of existing heat removal systems and have made them ideal working fluids for heat transfer enhancement in many industrial equipments including nuclear reactors (Buongiorno et al., 2009) and high-power electronic devices (Faulkner et al., 2003).

For the purpose of system design and performance assessment, a robust model capable of predicting heat transfer by boiling nanofluids is in great demand. Due to the near-molecular mixing (Wen et al., 2009) between the nanoparticles and the base liquid, a dilute nanofluid behaves hydro-dynamically like its pure base liquid and can be numerically treated as a single liquid phase despite the existence of two phases. This has allowed in the literature developing thermal-fluid dynamic models (Palm, Roy et al., 2006) for nanofluids based on computational fluid dynamics (CFD). For vapor-liquid two-phase flows of nanofluids with heat and mass transfer, our recent study (Li et al., 2014) demonstrated that the two-fluid model (Ishii, 1975) is still applicable. However, due to the specific phenomena observed boiling nanofluids such as surface modifications (Wen, Corr et al., 2011) and flow modifications (Domínguez-Ontiveros et al., 2010) which are not presented in nucleate boiling of pure liquids, the closure correlations/models of the two-fluid model have to be properly reformulated to account for the specific features induced by the existence of nanoparticles.

A comprehensive literature survey (Vafaei and Borca-Tasciuc, 2013) revealed that the forming of a porous layer of deposited nanoparticles on the heater surface, which is believed to be caused by evaporation of the liquid microlayer, is one of the common findings of most experimental studies on nucleate boiling of nanofluids. This porous layer not only changes the surface morphology and properties, but also alters the characteristics of bubble nucleation and departure, and is widely believed to be the essential cause of the dramatic boiling heat transfer performance of nanofluids.
Therefore, as proven in our recent study (Li et al., 2014), the key issue when formulating a theoretical model for nucleate boiling of dilute nanofluids is to characterize the surface modifications and the altered bubble nucleation behaviors.

Therefore in this study, new closure correlations were incorporated into the heat flux partitioning (HFP) model by Kurul and Podowski (1990) in order to capture the characteristics of heat and mass transfer on nanoparticle-deposited heater surfaces. Parametric studies were performed to analyze the effects of the improved surface wettability and altered surface roughness on bubble nucleation and departure. The HFP model was then incorporated as a boundary condition into the fully validated two-fluid model for boiling flows (Tu and Yeoh, 2002; Li et al., 2007) and 2-D numerical computations were conducted using the commercial CFD code CFX 4.4. The numerical results were compared against both the experimental data available in the literature and the classic Rohsenow pool boiling correlation.

### 3.1.2 The Heat Flux Partitioning (HFP) Model

Although the morphology and properties of the heater surface have been significantly changed by the deposited nanoparticles, the boiling heat transfer mechanisms involved on a nano-coated surface are believed to keep unchanged as those on a clean surface. Therefore, the HFP model proposed by Kurul and Podowski (1990) is still mechanistically applicable to nucleate boiling of nanofluids. According to the HFP model, the heat flux from a heater surface is transferred into the fluids through three mechanisms, namely the evaporation, quenching and convection mechanisms by

\[
q = q_e + q_q + q_c
\]

where, \(q_e\), \(q_q\) and \(q_c\) represent the heat flux components transferred by evaporation, quenching and convection, respectively.

\[
q_e = \frac{\pi}{6} d_{bh}^3 \rho_e f N_a h_f
\]

\[
q_q = \frac{2}{\sqrt{\pi}} f A_q \sqrt{r_w \lambda_l \rho_t c_p L} (T_w - T_l)
\]

\[
q_c = A_c St \rho_t c_p u_1 (T_w - T_l)
\]
where, $d_{bw}$, $f$, $N_a$, $t_w$, $A_c$ and $A_q$ are the bubble departure diameter, bubble departure frequency, active site density, bubble waiting time, the area fractions of the heater surface subjected to convection and quenching, respectively. Due to the inherent complexity of bubble nucleation and departure, these parameters are generally formulated empirically or semi-empirically. Although a number of correlations are available in the literature and some of them have been fully validated for boiling of pure liquids, however, their applicability for nanofluids is still open to question. For the purpose of effective modeling, the nucleate boiling parameters have to be carefully formulated.

### 3.1.2.1 The Active Site Density

According to the classic Bankoff bubble nucleation mechanism, the availability of cavities on a heater surface for bubble nucleation is strongly affected by the surface microstructures and wettability. Based on this mechanism and a cone cavity assumption, Yang and Kim (1988) correlated the active site density to the surface microstructures and liquid contact

$$N_a = N \int_0^{\beta/2} f(\beta) d\beta \times \int_{r_{c_{\text{min}}}}^{r_{c_{\text{max}}}} f(r_c) dr_c$$

(3.5)

where, $N$ is the total number of possible nucleation sites available on a unit heater surface area, $f(\beta)$ and $f(r_c)$ are the probability density functions for the cone angle ($\beta$) and cavity mouth size ($r_c$), respectively. The key issue when applying the Yang-Kim correlation is to provide statistical parameters for the surface microstructures ($f(\beta)$ and $f(r_c)$), which depends on the heater material and surface polishing and have to be determined experimentally. Unfortunately, due to the diversity and inherent complexity of realistic heater surfaces, it’s anything but an easy job to formulate a universal active site density correlation based on Equation 3.5.

As a simplification, the active site density has been widely correlated to the wall superheat and some other parameters such as the liquid contact angle and the surface roughness. Among them, the correlations proposed by Benjamin and Balakrishnan (1997), Wang and Dhir (1993) and Basu et al. (2002) are highly regarded in terms of accuracy for nucleate boiling of water (Hibiki and Ishii, 2003). However, a recent comparison (Li et al., 2014) of these correlations against the experimental data of
aqueous nanofluids proved that they are actually not applicable to nanofluids. This is perhaps due to the fact that they are empirical and limited to pure liquids. For nanofluids, the active density needs to be reformulated so that the effects impacted by the nanoparticles could be taken into account.

It is widely believed that the deposited nanoparticles affect bubble nucleation through two ways (Kim et al., 2007). Firstly, they alter the total number of sites available for bubble nucleation by changing the microstructures of the heater surface. Secondly, the deposited nanoparticles largely improve the wettability of the heater surface, which causes a part of the nucleation sites being flooded by the liquid and cannot be activated. Therefore, it is crucial to take both the morphology and property modifications into account when modelling the active site density of nanofluids.

In order to describe the effects of nanoparticle deposition on bubble nucleation, Ganapathy and Sajith (2013) proposed a semi-analytic correlation for the active site density based on the Benjamin-Balakrishnan correlation (Benjamin and Balakrishnan, 1997), in which both the wettability enhancement and the nanoparticle size relative to the surface roughness were considered

\[ N_a = 218.8 \frac{1}{\gamma} P^{1.63} \left( 14.5 - 4.5 \left( \frac{R_a P}{\sigma} \right) + 0.4 \left( \frac{R_a P}{\sigma} \right)^2 \right) \beta^{-3} \left( \frac{R_a}{d_p} \right)^{-0.5} \Delta T_{sup}^{0.4} \]  

(3.6)

where \( P, R_a \) and \( d_p \) stand for the pressure, average surface roughness and nanoparticle diameter, respectively. \( \gamma \) is the wall-liquid interaction parameter determined by the surface and liquid materials and \( \beta \) is the surface wettability improvement parameter defined by

\[ \beta = \frac{1 - \cos \theta}{1 - \cos \theta^*} \]

(3.7)

where \( \theta \) and \( \theta^* \) are the liquid contact angles on the nanocoated surface and clean surface, respectively. Comparing Equation 3.6 with the Benjamin-Balakrishnan correlation (Benjamin and Balakrishnan, 1997), it is clear that the term \( \beta^{-3} \) accounts for the improved surface wettability and \( (R_a/d_p)^{-0.5} \) describes the change of surface roughness.

Equation 3.6 was plotted in Figure 3.1 and was compared against the experimental data of nanofluids by Gerardi et al. (2011). Figure 3.1 indicated that the Ganapathy-
Sajith correlation predicts a decreased active site density with improved surface wettablity, which agrees phenomenologically with the experimental observations (Kim et al., 2007). Figure 3.1 also indicated that Equation 3.6 agrees well with the experimental data when \( \theta = 30^\circ \), which also agrees with Kim et al.’s experimental measurements (2007) that the liquid contact angle of on a nanocoated surface was in the range of \( 8^\circ \sim 36^\circ \). In addition, Equation 3.6 predicted that the active site density decreases monotonously with increased nanoparticle size under a given surface roughness. However, this doesn’t agree with the survey conclusion by Narayan et al. (2007) and Das et al. (2008) that heat transfer by nanofluids is deteriorated when \( R_a/d_p \) approaches 1.0, otherwise heat transfer is enhanced as \( R_a/d_p \) is away from 1.0. They proposed that when \( R_a/d_p \) is near 1.0, deposited nanoparticles reset in the cavities on the heater surface and reduce the active site density (Narayan et al., 2007). Otherwise when the surface roughness and particle size were apart away, more active site density would be created, especially when the nanoparticle size is smaller than the roughness, nanoparticles trapped in a big cavity can split it into two or more active nucleation sites and hence largely increase the heat transfer performance. Therefore, in order that the effects of particle size relative to the surface roughness could be effectively considered, Equation 3.6 was reformulated in this study by

\[
N_a = C_n \left( \frac{R_a P}{\sigma} \right)^{1.03} \left( 14.5 - 4.5 \left( \frac{R_a P}{\sigma} \right)^2 \right)^{-0.4} \left( \frac{R_a}{d_p} \right) \Delta T_{\text{sup}}^{3.0} \tag{3.8}
\]

\[
\frac{\xi}{d_p} = \begin{cases} 
0.275 \left( \frac{R_a}{d_p} \right)^{-1.2} & \text{if } R_a/d_p \leq 1.0 \\
0.275 + 0.7911 \left( R_a/d_p - 1 \right)^{0.68} & \text{if } R_a/d_p > 1.0
\end{cases} \tag{3.9}
\]

where, \( C_n \) is an empirical constant and \( C_n = 512 \) in this study.

In addition, as the heater surface is fully coated by the deposited nanoparticles, bubble nucleation no longer occurs on the original heater surface, but actually occurs on the layer of deposited nanoparticles, the wall-liquid interaction parameter in Equation 3.6 was therefore re-defined in this study by

\[
\gamma = \frac{\lambda_p c_{p,p}}{\sqrt{\lambda_r c_{p,r}}} \tag{3.10}
\]
Figure 3.1: Comparison of the Ganapathy-Sajith correlation (Ganapathy and Sajith, 2013) against experimental data (Gerardi et al., 2011): (a) effect of the liquid contact angle; (b) effect of nanoparticle size.

For the purpose of comparison, the active site density correlations recently proposed by Li et al. (2014b) and by Hibiki and Ishii (2003) were also included in this study. The Li correlation (Equation 3.11) was fitted using the nanofluid experimental data. The Hibiki-Ishii correlation (Equation 3.12) was based on Yang and Kim (1988) (Equation 3.5) and was originally proposed for pure liquids. However, Equation 3.12 was still included in this study as it was stated to be applicable to a wide parametric
range (0.101 MPa ≤ P ≤ 19.8 MPa, 5° ≤ θ ≤ 90°, and 1×10^4 ≤ n ≤ 1.51×10^{10} sites/m^2) which actually covers most conditions in nucleate boiling of nanofluids.

\[
N_a = 1.206 \times 10^4 (1 - \cos \theta) \Delta T^{2.06}_{sup} \tag{3.11}
\]

\[
N_a = \bar{N}_a \left(1 - \exp \left(-\frac{\theta^2}{8\varsigma^2}\right)\right) \exp \left(f \left(\rho^+\right) \frac{f}{R_e}\right) - 1 \tag{3.12}
\]

Where

\[
R_e = \frac{2\rho_v h_{fl} \Delta T_{sup}}{\sigma \rho_v h_{fl} \Delta T_{sup}} \tag{3.13}
\]

\[
f \left(\rho^+\right) = -0.01064 + 0.48246 \rho^+ - 0.22712 \rho^{+2} + 0.05468 \rho^{+3} \tag{3.14}
\]

\[
\rho^+ = \log \frac{\rho_l - \rho_v}{\rho_v} \tag{3.15}
\]

Empirical constants in Equation 3.12 are \(\bar{N}_a = 4.72 \times 10^5\) sites/m^2, \(\varsigma = 0.772\) rad and \(l' = 2.50 \times 10^{-6}\) m.

### 3.1.2.2 Other Nucleate Boiling Parameters

At first, the bubble departure diameter is another important nucleate boiling parameter needing in-depth study and further formulation. Although a number of correlations have been proposed since 1930s (e.g., the famous Fritz correlation), however, as proven by Kolev (2012) who conducted a comprehensive comparison of various bubble departure diameter correlations available in the literature against the experimental data of water published by different investigators, a universal correlation which fits most experimental data of pure liquids is still absent.

For nanofluids, the situation is even more challenging as quantitative studies on the bubble departure diameter are very rare. Considering the improved surface wettability in boiling nanofluids has a significant effect on the characteristics of bubble departure, Phan et al. (2009) proposed a new bubble departure diameter correlation by reformulating the Fritz correlation, in which the liquid contact angle is included.

\[
d_{bw} = 0.626977 \left(\frac{2+3 \cos \theta - \cos^3 \theta}{4}\right) \sqrt{\frac{\sigma}{g \left(\rho_l - \rho_v\right)}} \tag{3.16}
\]
Equation 3.16 predicts an increasing bubble departure diameter with improved surface wettability, which phenomenologically agrees with most experiments of nanofluids. However, it should be noted that for a given nanofluid, Equation 3.16 is correlated only to the liquid contact angle while other factors are ignored.

It’s still very challenging today to formulate a mechanistic correlation or model of the bubble departure diameter even for pure liquid, without saying the so many novel features in nanofluids. As a simplification, a polynomial correlation was obtained for the bubble departure diameter in this study by fitting Gerardi et al.’s data (2011).

\[
d_{bw} = -1.91 \times 10^{-3} + 4.21125 \times 10^{-4} \Delta T_{sup} - 1.70945 \times 10^{-5} \Delta T_{sup}^2 + 2.03938 \times 10^{-7} \Delta T_{sup}^3
\]  

(3.17)

The bubble departure frequency has been widely observed in various experiments to decrease with increasing bubble departure diameter, for both pure liquids (Situ et al., 2008) and nanofluids (Gerardi et al., 2011). It’s physically reasonable that a larger bubble needs a longer time to grow, which leads to a prolonged bubble period and a reduced bubble departure frequency. The bubble departure frequency is generally correlated to the bubble departure diameter in the form of

\[
f = 1/d_{bw}^k
\]  

(3.18)

However, the index \( k \) takes different values in various correlations. For example, \( k = 1/2 \) in the Cole correlation (Cole, 1960) (Equation 3.19), \( k = 1.5 \) in the Stephan correlation (Stephan, 1992) (Equation 3.20) and \( k = 2 \) in the Hatton-Hall correlation (Hatton and Hall, 1966) (Equation 3.21). The applicability of these correlations to nanofluids and determination of \( k \) will be discussed later.

\[
f = C_f \sqrt{\frac{4g(\rho_f - \rho_l)}{3d_{bw} \rho_l}}
\]  

(3.19)

\[
f = \frac{1}{\pi} \sqrt{\frac{g}{2d_{bw} \rho_f}} \left(1 + \frac{4\sigma}{d_{bw}^2 \rho_f g}\right)
\]  

(3.20)

\[
f = 284.7 \frac{\lambda_i}{d_{bw}^2 \rho_f c_{p,l}}
\]  

(3.21)

The remaining nucleate boiling parameters \( t_w \), \( A_c \) and \( A_q \) are defined in the same way as that in our previous studies (Tu and Yeoh, 2002; Li et al., 2009).
\[ t_w = 0.8/f \quad (3.22) \]

\[ A_q = 1 - A_c = N_u K \frac{\pi d_{bw}^2}{4} \quad (3.23) \]

### 3.1.3 Numerical procedures

The aforementioned HFP model was solved using an iterative bisection algorithm. Since the heat and mass transfer on a heater surface is not physically independent of the bulk flow field, the HFP model was incorporated as a boundary condition into the two-fluid model governing mass, momentum and energy conservation of the two phases. The two-fluid model for nucleate boiling could be written in a form of the generic scalar advection-diffusion equation for the general two-phase flow variable \( \phi \):

\[
\frac{\partial}{\partial t} \left( \alpha_i \rho_i \phi_i \right) + \nabla \cdot \left( \alpha_i \rho_i \vec{U}_i \phi_i - \Gamma_i \nabla \phi_i \right) = \alpha_i S_i + c_{ij} \left( \phi_j - \phi_i \right) + (m_{ij} \phi_j - m_{ji} \phi_i) \quad (3.24)
\]

Details of the two-fluid model have been extensively highlighted in our previous works (Tu and Yeoh, 2002; Li et al., 2007) and will not be repeated here. Due to the continuous nanoparticle deposition as a result of microlayer evaporation, nucleate boiling of nanofluids is strictly a transient process as reported by many investigators (Kim et al., 2007). However, the experimental observations by Okawa et al. (2012) demonstrated that as the heater surface was fully coated by nanoparticles, the surface morphology and properties as well as the heat transfer performances did not change any further despite the ongoing nanoparticle deposition. The duration of the transient initial stage was generally short (e.g. roughly 20 minutes according to Okawa et al. (2012) and nucleate boiling of nanofluids was predominantly characterized by a quasi-steady state. Therefore, this study ignored the transient initial stage by excluding the time derivative (the first term on the left-hand-side of Equation 3.24) and focused only on the steady stage.

Then pool boiling of dilute aqueous nanofluids in a cylindric pool containing a small circular heater located at the centre of its bottom was simulated using the aforementioned models. The pool (400 mm in diameter and 200 mm in height, as illustrated in Figure 3.2) was created much larger than the heater (20 mm in diameter)
so that the flow and heat transfer in the vicinity of the heater surface is free from the edge effects. Due to the axisymmetric distribution of the flow field, a two-dimensional computational domain (200 mm-radius \( \times \) 200 mm-height) was built. The domain was then discretized using hexahedral structured meshes. Mesh sensitivity test proved that mesh independence was achieved at 300 (in radius) \( \times \) 150 (in height) cells since a further increase of mesh density to 400\( \times \)200 cells just caused a negligible change (less than 0.5\%) in the heat transfer coefficient.

Since the addition of a small amount (less than 0.1 vol\%) of nanoparticles into the base liquid has only a negligible effect on its physical properties (Kim et al., 2007), the property parameters (e.g. viscosity, thermal conductivity, specific heat, saturation temperature and latent heat) of pure water were employed for the liquid phase. In fact, our recent study (Li et al., 2014b) has demonstrated that modification of the liquid properties considering the existence of nanoparticles in the base liquid has only a nearly invisible effect (less than 0.5\%) on the two-fluid model predictions. Therefore, it’s safe to ignore the liquid property changes induced by the existence of nanoparticles. An atmospheric pressure condition was applied at the pool surface. Vapor release at the pool surface was modeled by introducing a degassing boundary, which acted as a vapor sink depending on the rising velocity of vapor bubbles and the vapor volume fraction.

Numerical computations were performed using the commercial CFD code CFX-4. Convergence was achieved with 5000 iterations when the residual of the continuity equation of the liquid phase dropped to less than \( 1 \times 10^{-5} \). A number of computations
were performed to analyze the effects of surface wettability and nanoparticle material and size on the heat transfer performance, as discussed in the following sections.

3.1.4 Results and Discussion

3.1.4.1 Comparison against experimental data

The HFP model incorporated with new closure correlations was compared against the experimental data of dilute SiO$_2$/water nanofluid (0.1 vol%) by Gerardi et al. (2011). Based on the experimental measurements by Kim et al. (2007), $\theta^* = 79^\circ$ and $\theta = 22^\circ$ were selected for the liquid contact angles on the clean and nanoparticle-deposited heater surfaces during the computations, respectively.

![Graph showing comparison of active site density prediction against experimental data](Gerardi et al., 2011)

Figure 3.3: Comparison of active site density prediction against experimental data (Gerardi et al., 2011).

As demonstrated in our previous study (Li et al., 2014b), the active site density modeling has the most significant effect on the overall prediction using the HFP model. Therefore, the active site density formulation was firstly tested in this study. The active site density predicted by the HFP model incorporated with different active site density correlations was compared against the experimental data in Figure 3.3. It revealed that the Hibiki-Ishii correlation (Equation 3.12) largely under-predicted the active site density in nanofluids, although it has been fully validated for nucleate boiling of pure liquids. This was perhaps due to the fact that the empirical constants in Equation 3.12...
were based on experimental data of pure liquids where surface modifications did not exist. In contrast, the new active site density correlation developed in this study (Equation 3.8) and that fitted by Li et al. (2014) (Equation 3.11) achieved good agreements with the experimental data. However, Equation 3.8 predicted a larger increasing speed of the active site density than Equation 3.11 with the improved wall temperature.

The predicted bubble departure diameter was compared against the experimental data of nanofluid in Figure 3.4. According to Kolev’s literature survey (2012), the bubble departure diameter in boiling water under the atmospheric pressure is strongly affected by the wall temperature. It increases at first with the improved wall temperature and reaches its maximum at $\Delta T_{sup} = 15\sim20$ K. The bubble departure diameter then decreases as the wall temperature keeps increasing. For nucleate boiling of SiO$_2$/water nanofluids, the same tendency was observed by Gerardi et al. (2011) and the turning point also appeared at $\Delta T_{sup} = 15\sim20$ K (Figure 3.4). This evolution was successfully predicted by the new bubbled departure diameter correlation of this study (Equation 3.17). Besides the bubble departure diameter correlation by Phan et al. (2009) (Equation 3.16) and Equation 3.17, the widely used correlations proposed by Stephan (1992) and by Lemmert and Chwala (1977) were also included in this study. However, obvious deviations were observed with these correlations. In fact, the bubble departure diameter is affected by a number of factors including the surface and liquid properties, system pressure and heater surface temperature (Gerardi et al., 2011; Kolev, 2012). Unfortunately, a comprehensive model/correlation which takes most of these factors into account is still not available. Further research is in urgent demand in this area.

The bubble departure frequency predicted by various correlations was compared against the experimental data (Gerardi et al., 2011) in Figure 3.5. It was found that for aqueous nanofluids, $k = 1/2$ achieved the closest developing profile with the experimental data as the wall temperature increased. Therefore in the following sections of this study, the Cole correlation (Cole, 1960) (Equation 3.19) was utilized to calculate the bubble departure diameter. For the purpose of model calibration, a coefficient $C_f$ was added and it was found that when $C_f = 0.5$ Equation 3.19 agreed well with the experimental data of SiO$_2$/water nanofluids by Gerardi et al. (2011).
As the nucleate boiling parameter correlations had been validated, the predicted wall superheat of the heater surface under various heat flux was compared against the experimental data (Gerardi et al., 2011) in Figure 3.6. Since nanoparticle deposition has the most significant effects on bubble nucleation (Li et al., 2014b), the results yielded from the HFP model incorporated with various active site density correlations (the Li correlation (Equation 3.11) and the Hibiki-Ishii correlation (Equation 3.12)) were also included in Figure 3.6 for the purpose of comparison. It demonstrated that the HFP
model incorporated with the new active site density correlation (Equation 3.8) achieved the best agreement with the experimental data in the whole heat flux range (0~500 kW/m²). As expected, the HFP model largely over-predicted the surface temperature when the Hibiki-Ishii correlation (Equation 3.12) was incorporated as Equation 3.12 largely under-predicted the active site density in nanofluids (see Figure 3.3).

Figure 3.6: Predicted wall superheat vs. experimental data (Gerardi et al., 2011).

3.1.4.2 Prediction of pool boiling experimental data of aqueous-oxide nanofluids

A number of metallic and non-metallic materials could be used to prepare nanofluids (Vafaei and Borca-Tasciuc, 2013). Despite the diversity, oxides are expected to be promising for heat transfer applications thanks to their stable physical and chemical properties and excellent environment compatibility (Buongiorno and Hu, 2009). Therefore, this study focused on aqueous-oxide nanofluids. Physical properties of several widely used oxides as summarized by Vafaei and Borca-Tasciuc (2013) and their wall-liquid interaction parameters with saturated water under the atmospheric pressure are listed in Table 3.1. Theoretically, the nanoparticle material would affect the boiling heat transfer performance through altering the wall-liquid interaction parameter (Equation 3.10).

For a heater boiled in a given aqueous-oxide nanofluid, Kim et al. (2007) found in their experiments that the surface wettability was affected by the nanoparticle
concentration, as shown in Table 3.2. Therefore, the effect of particle concentration could be taken into account through the liquid contact angle.

In terms of Equation 3.8, the active site density increases with the decreased surface wettability or decreased wall-liquid interaction parameter. This means that within the parametric range specified in Table 3.1 and 3.2, the best heat transfer performance appears at $\theta = 43^\circ$ and $\gamma = 0.931$ while the lowest heat transfer coefficient happens at $\theta = 11^\circ$ and $\gamma = 3.788$. The predicted upper and lower limits of boiling curves were compared against the experimental data available in the literature (Vassallo et al., 2004; Bang and Chang, 2005; Chopkar et al., 2008; Coursey and Kim, 2008; Suriyawong and Wongwises, 2010; Harish et al., 2011; Huang et al., 2011; Ahmed and Hamed, 2012; Shahmoradi et al., 2013; Shoghl and bahrami, 2013) in Figure 3.7. The experimental data selected for comparison were limited to pool boiling of dilute aqueous-oxide nanofluids under the atmospheric pressure. Most nanofluids presented in Figure 3.7 had a concentration lower than 0.1% by volume. Figure 3.7 demonstrated that over 95% of these experimental data fell within the range defined by the upper and lower boiling curves.
Taylor and Phelan (2009) once conducted a comprehensive comparison of pool boiling experimental data of aqueous nanofluids available in the literature against the classic Rohsenow correlation (Equation 3.25).

\[
q = \mu \lambda h_{fg} \sqrt{\frac{(\rho_l - \rho_v) g}{\sigma}} \left( \frac{1}{C_{sf}} \right)^{0.33} \left( \frac{1}{Pr} \right)^{0.33} \left( \frac{c_p \Delta T_{sup}}{h_{fg}} \right)^{0.33}
\]  

(3.25)

They found that almost all the experimental data could be fitted to the Rohsenow correlation with the surface constant \( C_{sf} \) varying from 0.0065 to 0.018. This indicated that the surface modifications induced by nanoparticle deposition is the major factor responsible for the dramatic heat transfer performance since the surface constant \( C_{sf} \) is correlated only to the surface conditions. However, Taylor and Phelan’s comparison (2009) was conducted in a relatively narrow parametric range (wall superheat less than 15 K) and the Rohsenow correlation was not fully assessed. In this study, the Rohsenow correlation was compared against the HFP model and the experimental data in a wider parametric range, as illustrated in Figure 3.7, which demonstrated that for a wider wall superheat range (up to 45 K), the HFP model provided a better prediction than the Rohsenow correlation. Furthermore, as pointed by Taylor and Phelan (2009), it is not practical to predict nucleate boiling of nanofluids using the Rohsenow correlation since the surface constant \( C_{sf} \) needs to be determined experimentally. Comparatively, the HFP model of this study provides a more feasible approach to predict the heat transfer by boiling nanofluids and its applicable range is much wider.

### 3.1.4.3 Further Discussion

As the dramatically changed heat transfer performance of boiling nanofluids is due to the surface modifications induced by nanoparticle deposition, it is expected that the properties of a nanoparticle-deposited heater surfaces as well as the characteristics of bubble nucleation may be different depending on the material, size and concentration of the nanoparticles. The deposited nanoparticles affect nucleate boiling mainly through three ways: At first, the nanoparticle material affects the wall-liquid interaction parameter and the active site density through Equation 3.10 (see Table 3.1). Secondly, the wettability of a nanoparticle-deposited surface is directly affected by the nanoparticle material and concentration (see Table 3.2). Finally, as proven by Narayan...
et al. (2007) and Das et al. (2008), the nanoparticle size is another important parameter affecting the heat transfer performance on a nanoparticle-deposited heater surface, since the deposited nanoparticles may increase or decrease the active site density depending on their size relative to the roughness of the clean heater surface.

### Table 3.1 Physical properties of the nanoparticle materials and water.

<table>
<thead>
<tr>
<th>Material</th>
<th>Density (kg/m³)</th>
<th>Thermal conductivity (W/(m K))</th>
<th>Specific heat (J/(kg K))</th>
<th>γ (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Saturated water</td>
<td>958</td>
<td>0.679</td>
<td>4216</td>
<td>1.000</td>
</tr>
<tr>
<td>SiO₂</td>
<td>2410</td>
<td>1.4</td>
<td>705</td>
<td>0.931</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>3490</td>
<td>25.0</td>
<td>451</td>
<td>3.788</td>
</tr>
<tr>
<td>ZrO₂</td>
<td>5570</td>
<td>2.2</td>
<td>480</td>
<td>1.465</td>
</tr>
<tr>
<td>ZnO</td>
<td>5606</td>
<td>3.2</td>
<td>580</td>
<td>1.948</td>
</tr>
<tr>
<td>TiO₂</td>
<td>4010</td>
<td>8.3</td>
<td>690</td>
<td>2.894</td>
</tr>
</tbody>
</table>

### Table 3.2 Liquid contact angle on heater surfaces boiled in different nanofluids (Kim et al., 2007).

<table>
<thead>
<tr>
<th>Nanofluids</th>
<th>0.001vol%</th>
<th>0.01vol%</th>
<th>0.1vol%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al₂O₃/water</td>
<td>14°</td>
<td>23°</td>
<td>40°</td>
</tr>
<tr>
<td>ZrO₂/water</td>
<td>43°</td>
<td>26°</td>
<td>30°</td>
</tr>
<tr>
<td>SiO₂/water</td>
<td>11°</td>
<td>15°</td>
<td>21°</td>
</tr>
</tbody>
</table>

Computations were conducted to analyze the above factor individually, which would certainly contribute to an in-depth understanding of the mechanisms associated with nucleate boiling of nanofluids. During the computations of testing a certain factor (e.g. θ), the other two parameters (e.g. γ and $R_a / d_p$) were kept constant. The computations were conducted in the heat flux range of 10 – 500 kW/m² and the predicted active site density under various conditions was illustrated in Figure 3.8. The results demonstrated that the liquid contact angle has the most significant effect on the active site density as θ increasing from 8° to 36° caused a threefold increase in the active site density. The nanoparticle material also had a significant effect on the characteristics of bubble nucleation. Comparatively, the nanoparticle size had the least impact, although an increasing nanoparticle size firstly caused a decrease and then an increase in the active site density, which was consistent with the summary by Narayan et al. (2007) and Das et al. (2008).
The HFP model presented in this study made it possible to predict nucleate boiling heat transfer of nanofluids in terms of the nanoparticle material and size, heater surface microstructure and properties. This actually provided a mechanistic description of heat and mass transfer on nanoparticle-deposited heater surfaces. Figure 3.8 indicated that as long as the surface morphology, the nanoparticle size and the liquid contact angle on a nanoparticle-deposited heater surface could be effectively characterized, the heat transfer performance as well the nucleate boiling parameters of a given nanofluid could be quantitatively predicted. Unfortunately, due to the inherent complexity, a comprehensive experimental study considering all of these factors is still absent from the literature. Fundamental studies aimed to characterizing the microstructures and properties of heater surfaces coated with nanoparticles are strongly recommended.

![Figure 3.8: Effects of liquid contact angle, particle size and nanoparticle material on bubble nucleation.](image)

### 3.1.5 Conclusions

Compared with pure liquids, dilute nanofluids present similar hydro- and thermodynamic properties. However, due to the surface modifications induced by nanoparticle deposition which were not observed in nucleate boiling of pure liquids, nanofluids present dramatically changed bubble nucleation characteristics and heat transfer performance. Therefore, it’s crucial to characterize the surface modifications and their
effects on bubble nucleation when modeling nucleate boiling of nanofluids. In this study, new correlations of the nucleate boiling parameters were incorporated into the classic HFP model (Kuru and Podowski, 1990). Numerical computations and parametric study were performed to analyze the effects of nanoparticle material, size and concentration. Conclusions arising from this study are as follows:

(1) Through incorporating new closure correlations to account for the effects of surface modifications on the characteristics of bubble nucleation and departure, the HFP model achieved a satisfactory agreement with most experimental data of nucleate boiling of aqueous-oxide nanofluids available in the literature. The improved HFP model also provided a more feasible and mechanistic approach than the classic Rohsenow correlation to predict nucleate boiling of nanofluids.

(2) The surface wettability enhancement induced by nanoparticle deposition, among the other parameters (θ, γ and Ra / dp) investigated in this study, had the most significant effect on bubble nucleation on the nanoparticle-deposited heater surface.
3.2 A Theoretical Model Considering the Nanoparticle Brownian Motion in Liquid Microlayer

Abstract:

The forming of a porous layer of deposited nanoparticles on the heater surface is one of the unique phenomena in nucleate boiling of nanofluids. As the deposition of nanoparticles is induced by the evaporation of liquid microlayer, the average nanoparticle concentration in the microlayer is much higher than that in the bulk liquid. Therefore, the Brownian motion of the nanoparticles in microlayer may play an important role in dissipating heat from the heater surface. In this study, a new heat flux partitioning (HFP) model was proposed, in which a new heat flux component was incorporated to account for the heat transfer by the nanoparticle Brownian motion in liquid microlayer. The new heat flux component was formulated based on the latest experimental and theoretical research outcomes of microlayer evaporation. Comparison of the numerical results against the experimental data available in the literature proved that the new HFP model performs better than the classic HFP model. This study also demonstrated that the importance of nanoparticle Brwonian motion is mainly controlled by the applied heat flux as it directly affects the number density of active sites on the heater surface. Finally, the effects of nanoparticle concentration, size and materials were also analyzed.

3.2.1 Introduction

Nanofluids are colloidal dispersions of nano-sized particles in common base liquids. Due to their enhanced properties associated with heat transfer and the promising prospects of industrial applications, nanofluids have been attracting an increasing number of investigations (Wen et al., 2009). Following the study pioneered by Das et al. (2003a), heat transfer by nucleate boiling of nanofluids has been intensively studied, mainly through experimental approaches. According to the literature surveys (Jacqueline et al., 2011; Vafaei and Borca-Tasciuc, 2013), two common findings have been throw light on: (i) the significantly enhanced critical heat flux (CHF) and, (ii) the forming of a porous layer of deposited nanoparticles on the heater surface. These
phenomena were observed in almost all the experiments, even in those using dilute nanofluids with extremely low nanoparticle concentrations (Kim et al., 2007). For dilute nanofluids, numerous measurements (Kim et al., 2007; Kwark, 2009) have proven that their properties including the saturation temperature, surface tension, thermal conductivity and viscosity are negligibly different from those of their pure base liquids. Thus, the dramatically enhanced CHF is attributed not to the negligibly changed liquid properties, but exclusively to the surface modifications induced by nanoparticle deposition (Vafaei and Borca-Tasciuc, 2013).

In recent years, some efforts (Li et al., 2014a; Li et al., 2014b) have been devoted to develop predictive models for nucleate boiling of dilute nanofluids (typically with concentrations lower than 0.1 vol%) based on the heat flux partitioning (HFP) model (Kurul and Podowski, 1990). The effects of nanoparticles on the liquid properties were generally neglected in the models due to the aforementioned reasons, while focus was put mainly on the surface modifications and their effects on bubble dynamics (Li et al., 2014a; Li et al., 2014b). Through incorporating the active site density correlation of Ganapathy and Sajith (2013) and the bubble departure diameter correlation of Phan et al. (2009), the improved HFP model (Li et al., 2014a; Li et al., 2014b) achieved a better agreement with the experimental data available in the literature than the classic HFP model by Kurul and Podowski (1990).

However, an important mechanism may have been ignored. As to the forming of porous layers, Kim et al. (2007) and Kwark (2009) proved that the deposition of nanoparticles is caused by the evaporation of liquid microlayer. As illustrated in Figure 3.9, they proposed that when a bubble grows, the evaporating microlayer underneath the bubble leaves behind nanoparticles concentrating in it. The nanoparticles then adhere to the heater surface when the microlayer is completely vaporized. This indicates that within the bubble growth time, the concentration of nanoparticles in the microlayer would keep increasing from the bulk value up to 100%. Therefore, the time-averaged nanoparticle concentration in the microlayer would be much higher than the bulk value. According to a literature survey by Wang and Mujumdar (2007), the thermal conductivity enhancement of nanofluids at the atmospheric temperature could be as high as 60% when the nanoparticle concentration increased up to 5 vol%. In
addition, Das et al. (2003b) found that the effective thermal conductivity of nanofluids is a strongly increasing function of the temperature, much more considerable than that of pure liquids.

In fact, the enhanced thermal conductivity of nanofluids has been widely recognized and intensively studied. Since Jang and Choi (2004) attributed the dramatically improved thermal conductivity of nanofluids, for the first time, to the Brownian motion of nanoparticles in the liquid, this viewpoint has been widely accepted and a number of theoretical models for predicting the effective thermal conductivity of nanofluids have been proposed (Wang et al., 2003; Koo and Kleinstreuer, 2005; Pil Jang and Choi, 2007; Murshed et al., 2009). According to these models, the heat transfer due to nanoparticle Brownian motion increases with the nanoparticle concentration. Therefore, as the microlayer is a layer of superheated liquid with elevated nanoparticle concentration, the heat transfer contribution by the Brownian motion of nanoparticles may be significant.

In this study, a new HFP model was proposed. Apart from the heat flux partitioning components for convection, evaporation and quenching, a new component accounting for the heat transfer by nanoparticle Brownian motion in the microlayer was also incorporated in the new model. In addition, in consideration of the surface modifications induced by nanoparticle deposition, new correlations for the nucleate boiling parameters were carefully developed and selected. Numerical computations were then conducted using the both HFP models and their numerical results were compared against the experimental data available in the literature. Further computations were also conducted to analyze the factors affecting heat transfer by the nanoparticle Brownian motion.
3.2.2 Heat Flux Partitioning in Nucleate Boiling of Nanofluids

3.2.2.1 Heat Flux Partitioning in Boiling Nanofluids

For modeling of nucleate boiling of pure liquids, the classic HFP model developed by Kurul and Podowski (1990) has been widely recognized as a mechanistic approach. According to this model, the total heat flux $q$ applied at the heater surface could be partitioned into three components: the heat flux due to convection $q_c$, the heat flux due to evaporation $q_e$ and that due to quenching $q_q$.

$$q = q_c + q_e + q_q$$  \hspace{1cm} (3.26)

However, when nanoparticles exist in the liquid, the heat transfer mechanisms involved on the heater surface may be different. In this study, the HFP model was re-defined by adding a new component, $q_{bm}$, to model the heat transfer due to the nanoparticle Brownian motion. Therefore,

$$q = q_c + q_e + q_q + q_{bm}$$  \hspace{1cm} (3.27)

Equation 3.27 was termed as the new HFP model in the following sections. $q_c$, $q_e$ and $q_q$ were modeled in a mechanistic way by

$$q_c = \frac{\pi}{6} d_{bw}^3 \rho_c fN_a h_{f}$$  \hspace{1cm} (3.28)

$$q_q = \frac{2}{\sqrt{\pi}} fA_q \sqrt{f_w \lambda_h \rho c_p} (T_W - T_i)$$  \hspace{1cm} (3.29)

$$q_q = A_q h_c (T_W - T_i)$$  \hspace{1cm} (3.30)

where $d_{bw}$, $f$, $N_a$ and $t_w$ are the bubble departure diameter, bubble departure frequency, active site density and the bubble waiting time, respectively. $A_c$ and $A_q$ are the area fractions of the heater surface affected convection and quenching, respectively. $h_c$ is the single-phase convective heat transfer coefficient, which was modeled according to Krepper et al. (2007).

As aforementioned, the forming of a porous layer of deposited nanoparticles on the heater surface is one of unique features of nucleate boiling of nanofluids. This porous layer is believed to affect bubble nucleation mainly through two ways (Li et al., 2014b): (i) changing the surface microstructures and altering the number density of
cavities available for bubble nucleation and, (ii) largely improving the surface wettability which causes a portion of active sites being flooded. In consideration of the both effects, Ganapathy and Sajith (2013) proposed a semi-analytic correlation for the active site density in boiling nanofluids. In this study, their correlation (Ganapathy and Sajith, 2013) was further improved (Equation 3.31) to take into account the fact that heat transfer deterioration occurs when the ratio of surface roughness to nanoparticle size is near 1.0, otherwise heat transfer is enhanced as \( \frac{R_a}{d_{np}} \) is away from 1.0, as reported by Narayan et al. (2007) and Das et al. (2008).

\[
N_a = C_a \frac{1}{\gamma} P_r^{1.63} \left( 14.5 - 4.5 \left( \frac{R_p}{\sigma} \right) + 0.4 \left( \frac{R_p}{\sigma} \right)^2 \left( \frac{1 - \cos \theta}{1 - \cos \theta} \right)^{-3} \right)^{-0.4} \\
\xi \left( \frac{R_a}{d_{np}} \right)^{0.4} \Delta T_{np}^3
\]

(3.31)

where, \( \gamma \) is the liquid-wall interacting parameter. When the heater surface is fully coated by deposited nanoparticles, bubble nucleation no longer occurs on the original heater surface, but on the layer of deposited nanoparticles. Therefore, \( \gamma \) was defined by

\[
\gamma = \sqrt{\frac{\lambda_{np} P_{np} c_{p, np}}{\lambda_p c_{p, l}}}
\]

(3.32)

The term \( \xi \left( \frac{R_a}{d_{np}} \right) \) was added to describe the effects of nanoparticle size relative to the surface roughness

\[
\xi \left( \frac{R_a}{d_{np}} \right) = \begin{cases} 
0.275 \left( \frac{R_a}{d_{np}} \right)^{-1.2} & 0 < \frac{R_a}{d_{np}} < 1.0 \\
0.275 + 0.791 \left( \frac{R_a}{d_{np}} - 1 \right)^{0.68} & \frac{R_a}{d_{np}} \geq 1.0
\end{cases}
\]

(3.33)

In addition, the deposited nanoparticles were reported to change the force balance on the tri-phase contact line (Sefiane et al., 2008), thus may have a significant effect on the dynamic characteristics of bubble growth and departure. In fact, the bubble departure diameter is subjected to a number of factors. Due to the inherent complexity, a universal correlation for the bubble departure diameter is still absent even for pure liquids, let alone the novel features induced by nanoparticles. Although a few empirical or semi-empirical correlations have been proposed in recent years for the bubble departure diameter in boiling nanofluids or pure liquids boiling on nano-coated surfaces (e.g. the correlation of Phan et al., 2009), they are strictly limited to a certain applicable
range. As a simple approximation, the bubble departure diameter was correlated to the wall superheat in this study by fitting Gerardi et al.’s (2011) experimental data of silica/water nanofluids.

\[
d_{bw} = -1.91 \times 10^{-3} + 4.21125 \times 10^{-4} \Delta T_{sup} - 1.70945 \times 10^{-5} \Delta T_{sup}^2 + 2.03938 \times 10^{-7} \Delta T_{sup}^3 \quad 5 \text{ K} < \Delta T_{sup} < 35 \text{ K} \tag{3.34}
\]

Equation 3.34 is plotted in Figure 3.10. For the purpose of comparison, a couple of existing correlations (Stephan, 1992; Phan et al., 2009 and Lemmert and Chwala, 1977) were also plotted. Figure 3.10 indicates that the bubble departure diameter in boiling nanofluids increases firstly with the increased wall superheat. However, after hitting the peak value, the bubble departure diameter begins to decrease while the wall superheat is further improved. This is coincident with the trend in pure water, as summarized by Kolev (2012). However, yet no existing correlation could capture this trend. Comparatively, the bubble departure diameter correlation developed in this study (Equation 3.34), although not mechanistic, gave the best description of the effects of wall superheat on the bubble departure diameter.

![Figure 3.10: Bubble departure diameter as a function of the wall superheat.](image)

The other parameters in Equation 3.28 - 30 are defined by

\[
f = C_f \sqrt{\frac{4g(\rho_l - \rho_e)}{3d_{bw} \rho_l}} \tag{3.35}
\]
\[ t_w = 0.8/f \]  
\[ A_q = 1 - A_e = N_d K \frac{\pi d_{bw}^2}{4} \]  

### 3.2.2.2 Heat Transfer by Nanoparticle Brownian Motion in the Microlayer

This study focuses on dilute nanofluids in which the liquid properties are negligibly changed by the existence of nanoparticles. Therefore, the heat transfer due to nanoparticle Brownian motion was considered only in the microlayer. This heat transfer process could be equivalently treated as a thermal conduction across the microlayer. The heat transfer rate \( q_{bm} \) is thus defined by

\[ q_{bm} = N_d f \int_{0}^{t_{lm}} A_{lm} T_w - T_{sat} \lambda_{bm} \delta_{bm} \cdot dt \]

where, \( A_{lm} \) and \( \delta_{lm} \) are the area and thickness of the liquid microlayer, respectively. \( \lambda_{bm} \) is the equivalent thermal conductivity of nanoparticle Brownian motion. The microlayer acting time \( t_{lm} \) appears in Equation 3.38 to account for the fact that heat transfer by the nanoparticle Brownian motion acts only when the microlayer exists underneath the bubble.

Fundamental knowledge about the geometrical parameters (\( A_{lm} \) and \( \delta_{lm} \)) of liquid microlayer is vital to formulating an effective correlation of \( q_{bm} \). In fact, although it has been widely accepted that the growth of bubble on an active site is attributed to the evaporation of liquid microlayer, quantitative measurements and characterizations of liquid microlayer are still very rare. In recent years, a couple of experimental measurements were conducted by Gao et al. (2013), Jung and Kim (2014) and Utaka et al. (2013) using various cutting-edge technologies, with the aim to characterize the dynamic process of liquid microlayer evaporation. These experimental measurements, although have not contributed to a theoretical model for the liquid microlayer, have provided important experimental data to estimate \( A_{lm} \) and \( \delta_{lm} \) in Equation 3.38.

The experimental observations (Gao et al., 2013; Utaka et al., 2013; Jung and Kim, 2014; Utaka et al., 2014; Chen and Utaka, 2015) proved that the liquid microlayer presents strong transient characteristics as the bubble grows. A typical evolving process of the microlayer along with the increasing bubble size is shown in Figure 3.11 (Jung...
and Kim, 2014). It reveals that the microlayer is formed as soon as a bubble is nucleated, which is accompanied by the simultaneous appearance of a smaller dry spot around the active site. Therefore, the liquid microlayer has an annular geometry with an outer diameter of $d_{\text{lm}}$ and an inner diameter of $d_{\text{lm}}$. As the bubble grows, both the microlayer and the dry spot expand their sizes, resulting in an enlarged annular area until the outer diameter reaches its maximum $d_{\text{lm},\text{max}}$ (Stage I). Afterwards, the outer diameter begins to shrink while the inner diameter keeps increasing, thus leading to a reduced microlayer area (Stage II). Finally as the inner diameter increases up to the outer diameter, the microlayer is completely dried up. However, the bubble does not depart immediately after the microlayer depletion, but keeps growing while the dry spot turns to shrink shortly after the microlayer depletion (Stage III). When the tri-phase contact line comes back close to the active site, bubble departure occurs.

![Figure 3.11: Evolution of the microlayer sizes as the bubble grows (Jung and Kim, 2014).](image)

According to Figure 3.11, the area of microlayer is defined by

$$A_{\text{lm}} = \frac{\pi}{4} \left( d_{\text{lm}}^2 - d_{\text{lm}}^2 \right)$$  \hspace{1cm} (3.39)

Due to the inherent complexity, a theoretical model for predicting the microlayer diameters is still absent. However, the experimental observations by Jung and Kim (2014) revealed that for a given liquid contact angle and bubble growth stage (Figure 3.11), a larger bubble requires a larger contact area with the heater wall, resulting in a
larger microlayer (Jung and Kim, 2014). Therefore, the microlayer sizes could be simply correlated to the bubble diameter. As a further simplification in this study, both \( d_{lmo} \) and \( d_{lmi} \) were correlated to \( d_{bW} \) based on the experimental data given in Figure 3.11, thus the mean microlayer area averaged over the microlayer acting time \( t_{lm} \) was estimated by

\[
\bar{A}_{lm} = \pi \left( \int_0^{t_{lm}} d_{lmo}^2 \cdot dt - \int_0^{t_{lm}} d_{lmi}^2 \cdot dt \right) / t_{lm} = 0.0806d_{bW}^2
\]  

(3.40)

In addition, the experimental observations (Gao et al., 2013; Utaka et al., 2013; Jung and Kim, 2014; Utaka et al., 2014; Chen and Utaka, 2015) found that the cross section of the annular microlayer always takes a triangular shape, as shown in Figure 3.12. As the bubble grows, the angle between the microlayer-vapor interface and the heater surface keeps constant (Figure 3.12(a)) when the interface was moving away from the active site centre. By looking at a fixed radius location, a linear reduction of the microlayer thickness is observed (Figure 3.12(b)).

Figure 3.12 indicates that the microlayer has an uneven thickness. However, this uneven thickness in the order of microns could be safely ignored when compared with the width of the microlayer annulus which is in the order of millimeters. Therefore, an even microlayer thickness was assumed in this study, with the mean value estimated based on the experimental data of Jung and Kim (2014).
The equivalent thermal conductivity of nanoparticle Brownian motion $\lambda_{bm}$ in Equation 3.38 is modeled according to Jang and Choi (2007) by

$$\lambda_{bm} = C_1 \lambda_{np} \varphi_{lm} + C_2 \frac{d_{bl}}{d_{np}} \lambda_{l} Pr_{lm} \varphi_{lm} Re_{np}^2$$  \hspace{1cm} (3.41)

where, $C_1$ is the constant for considering the Kapitza resistance (Huxtable et al., 2003) ($C_1 = 0.01$) and $C_2$ is a proportion constant ($C_2 = 18 \times 10^6$) (Jang and Choi, 2007). $\varphi_{lm}$ is the volumetric concentration of nanoparticles in the microlayer. $d_{bl}$ is the diameter of base liquid molecules ($d_{bl} = 0.384$ nm for water) and $Re_p$ is the nanoparticle Reynolds number depending on the mean velocity of random Brownian motion.

$$Re_p = \frac{\overline{C}_{RM} d_{np}}{\nu_{l}},$$  \hspace{1cm} (3.42)

$\overline{C}_{RM}$ is the random motion velocity of nanoparticle. By assuming that a nanoparticle moves freely over a distance of the mean free-path of the base liquid $l_{bl}$, $\overline{C}_{RM}$ could be calculated by

$$\overline{C}_{RM} = \frac{\kappa T_{lm}}{3 \pi \mu d_{p} l_{bl}},$$  \hspace{1cm} (3.43)
where, \( \kappa \) is the Boltzman constant and \( T_{lm} \) is the average temperature of the microlayer. By assuming a linear temperature distribution across the micron-thick micolayer, the average temperature is estimated by \( T_{lm} = (T_W + T_{sat}) / 2 \).

In addition, for a given nanoparticle concentration in the bulk liquid \( \varphi_0 \), the nanoparticle concentration in the microlayer could be modeled by

\[
\varphi_{lm}(t) = \frac{\varphi_0}{(1 - \varphi_0)(1 - t/t_{lm}) + \varphi_0} \tag{3.44}
\]

and the mean nanoparticle concentration averaged over the microlayer acting time \( t_{lm} \) is

\[
\bar{\varphi}_{lm} = \frac{\int_0^{t_{lm}} \varphi_m(t) dt}{t_{lm}} = \frac{\varphi_0}{1 - \varphi_0} \frac{\ln \frac{1}{\varphi_0}}{t_{lm}} \tag{3.45}
\]

Equation 3.44 and 45 are plotted in Figure 3.13(a) and (b), respectively. Figure 3.13(a) indicates that during the initial stage of microlayer evaporation, the nanoparticle concentration increases very slowly. Then the concentrating process gradually speeds up and finally a sharp increase of the concentration is observed close to the complete evaporation. This indicates that the nanoparticle Brownian motion contributes to heat transfer mainly in the later stage of a microlayer acting time. In addition, a strong nonlinear relationship between the bulk concentration and the mean concentration in the microlayer is observed (Figure 3.13(b)). When the bulk concentration is low (less than 0.01 \( \text{vol}\% \)), the average concentration in the microlayer is not sensitive to the bulk value. However, with increasing bulk concentration (larger than 0.1 \( \text{vol}\% \)), a small increase in the bulk concentration would lead to a sharp increase of the mean concentration.
Figure 3. 13: Nanoparticle concentration in microlayer: (a) evolution of nanoparticle concentration in the microlayer; (b) mean nanoparticle concentration in microlayer vs. the bulk concentration.

The equivalent thermal conductivity of nanoparticle Brownian motion $\lambda_{bm}$ is plotted versus the liquid temperature in Figure 3.14, which indicates that $\lambda_{bm}$ increases linearly with the microlayer temperature. In addition, when the bulk concentration is low (<0.1 vol%), $\lambda_{bm}$ is negligibly small (less than 0.02–0.03 W/(mK)) when compared with that of the base liquid (0.68 W/(mK)). However, $\lambda_{bm}$ increases significantly with improved bulk concentration. As the bulk concentration reaches 0.1 vol%, $\lambda_{bm}$ is larger
than 0.2 W/(mK), which is of the same magnitude order with that of the base liquid and hence cannot be neglected.

Figure 3. 14: The equivalent thermal conductivity of nanoparticle Brownian motion.

### 3.2.3 Results and Discussion

The aforementioned model equations were solved using an iterative bisection algorithm. Pool boiling of aqueous nanofluids under the atmospheric pressure was predicted within the heat flux range of 10 - 500 kW/m\(^2\).

#### 3.2.3.1 Model Validation and Analysis of HFP Components

The new HFP model was validated against the experimental data of Gerardiet al.’s (2011), who studied the pool boiling of dilute SiO\(_2\)/water nanofluid using the infrared thermometry. During the computations, the model parameters (e.g., \(\varphi_0\), \(d_{np}\), \(R_a\) and \(\theta\)) and boundary conditions were carefully set up based on the experimental conditions. Among them, the bulk concentration of SiO\(_2\) nanoparticles was 0.1 vol\%, the average nanoparticle diameters was 34 nm and the liquid contact angle on nanoparticle-deposited heater surface was 21°. Since the roughness of the clean heater surface was not given by the authors (Gerardi et al., 2011), it was estimated according to the NIST technical notes (Vorburger and Raja, 1990) that the roughness of electro-polished and super-finished metal surfaces was generally in the range of 25~200 nm with an average value of 100 nm. Theoretically, the surface roughness affects the predicted results.
mainly through the parameter $\zeta(R_a/d_{np})$ (Equation 3.33). For a given wall roughness, different ratios of $R_a/d_{np}$ could be realized by using various $d_{np}$ values (Section 3.2). More detailed investigation on the effects of wall roughness relative to nanoparticle size on nucleate boiling of nanofluids is available in our recent study (Li et al., 2015).

For the purpose of comparison, computations were also conducted using the classic HFP model. Firstly, the active site density predicted by the both HFP models was compared against the experimental data (Gerardi et al., 2011), as shown in Figure 3.15. The both models give satisfactory predictions to the active site density, which proves the validity of Equation 3.31 for nucleate boiling of nanofluids. However, for a given heat flux, the new HFP model predicts a lower active site density, which is caused by the lower prediction of the wall temperature, as will be explained in the following sections.

![Figure 3.15: Prediction of the active site density.](Image)

The boiling curves yielded from the both HFP models were also compared against the experimental data (Gerardi et al., 2011), as shown in Figure 3.16. Obviously, after incorporating the component $q_{bm}$, the new HFP model achieves a better agreement with the experimental data than the classic HFP model. This is especially true when the applied heat flux is elevated. For a given heat flux, the new HFP model predicts a lower wall superheat, indicating a higher heat transfer coefficient. The lower wall superheat predicted by the new HFP model gives a good interpretation to its lower prediction of the active site density as shown in Figure 3.15.
Figure 3.16: Comparison of predicted pool boiling curves against the experimental data (Gerardi et al., 2011).

The proportions of the HFP components predicted by the both models are plotted versus the heat flux in Figure 3.17. When the heat transfer by nanoparticle Brownian motion is not considered, the classic HFP model predicts that the quenching mechanism plays a major role in removing heat from the heater surface (Figure 3.17(a)). Especially when the applied heat flux is high (500 kW/m²), the quenching mechanism plays a predominant role by removing over 90% of the total heat. This agrees well with the conclusion drawn by Končar et al. (2004) and Tu and Yoeh (2002) who investigated nucleate boiling of pure water where the heat transfer by nanoparticle Brownian motion does not exist. However, when the contribution by nanoparticle Brownian motion is included, the new HFP model predicts that the significance of the quenching mechanism is largely reduced (Figure 3.17(b)), although it still plays a major role in heat removal (around 70% at 500 kW/m²). Moreover, a further comparison between Figure 3.17(a) and (b) indicates that the inclusion of nanoparticle Brownian motion in the HFP model does not cause much change to the contribution of the convective and evaporation mechanisms.
Figure 3.17: Comparison of heat flux components by the models: (a) classic HFP model; (b) new HFP model.

Figure 3.17(b) indicates that the proportion of the heat removal by nanoparticle Brownian motion increases with the elevated heat flux and reaches up to 22% when the applied heat flux is 500 kW/m². According to Equation 3.38, the surface area of the microalayer and the equivalent thermal conductivity are the key factors determining the heat transfer rate by the nanoparticles in microlayer. In order to achieve a deeper insight of the role of nanoparticle Brownian motion, the evolution of the parameters ($n$, $\tilde{n}A_{bn}$ and $\lambda_{bm}$) with the heat flux is plotted in Figure 3.18. The new HFP model predicts that the increasing heat flux improves the surface superheat, which creates more active
sites on the heater surface and leads to a larger area fraction of microalyers. The area fraction is as high as 7% when the heat flux increases up to 500 kW/m². In addition, the elevated wall temperature intensifies the nanoparticles’ Brownian motion in the microlayer, which leads to an increased equivalent thermal conductivity. All these factors working together make the nanoparticle Brownian motion play an increasingly important role in removing heat from the heater surface.

![Microlayer parameters vs. heat flux.](image)

Therefore, it is evident that the significance of nanoparticle Brownian motion in nucleate boiling of nanofluids is strongly affected by the applied heat flux. As the heat flux is elevated, the heat transferred by nanoparticle Brownian motion may take a considerable proportion and thus cannot be ignored.

### 3.2.3.2 Analyses of the Influencing Parameters

As the dramatic heat transfer performances of nucleate boiling of nanofluids are attributed to the deposition of nanoparticles, it is reasonable to expect that the material, size and concentration of the nanoparticles may have significant effects on the characteristics of nucleate boiling of nanofluids. In order to quantify the effects of each factor individually, further computations were conducted. It should be noted that the nanoparticles not just induce a new heat transfer mechanism in the microlayer (Equation 3.38), more importantly, they alter the dynamics of bubble nucleation.
through altering the surface morphology and properties. The both aspects working together have contributed to the dramatic features of nucleate boiling of nanofluids. Therefore in this study, the nanoparticle parameters (e.g., material, size and concentration) were analyzed in terms of their simultaneous impacts on the Brownian motion and bubble nucleation. During the computations, some necessary approximations were made. For example, Kim et al. (2007) found in their experiments that the liquid contact angle on a heater surface fouled with oxide nanoparticles, which was in the range of 8~36 degrees, was subjected to a number of factors including the nanoparticle material and concentration, and the applied heat flux. Since these factors have not been fully characterized, a mean value of 21 degrees was employed for the liquid contact angle in all the computational cases (Kim et al., 2007).

Computations were firstly conducted with different bulk concentrations (0.001~0.1 vol%), while the rest conditions were kept consistent with those in Section 3.1. The predicted results are presented in Figure 3.19. The computations indicated that the heat transfer is slightly enhanced (avg. 6% in the heat transfer coefficient) as the bulk concentration increases from 0.001 vol% to 0.1 vol% (Figure 3.19(a)). As the active site density is significantly reduced (Figure 3.19(b)), it was believed that this enhancement is not contributed by the evaporation mechanism. On the other hand, a dramatic increase over 2 orders of magnitude is predicted with the equivalent thermal conductivity \( \lambda_{bm} \) (Figure 3.19(c)), which causes a significant increase in the \( q_{bm} \) component (Figure 3.19(d)). As shown in Figure 3.19(d), when the bulk concentration is low (0.001 vol%), the importance of nanoparticle Brownian motion is negligible. However, as the bulk concentration increases up to 0.1 vol%, a considerable proportion of the heat flux is removed by the nanoparticles. The elevated heat flux further enhances this significance so that over 22% of heat is removed by the nanoparticles when the applied heat flux reaches 500 kW/m\(^2\). Therefore, for nucleate boiling of dilute nanofluids, increasing the nanoparticle concentration could largely improve the heat transfer component through nanoparticle Brownian motion nonetheless it would reduce the heat removal by evaporation.
Figure 3. 19: Effects of the bulk concentration. (Note: SiO₂/water, nanoparticle size 34 nm, surface roughness 100 nm).

Figure 3. 20: Effects of the nanoparticle size. (Note: 0.1 vol% SiO₂/water, surface roughness 100 nm).

The computational results yielded from different nanoparticle sizes are shown in Figure 3.20. Figure 3.20(a) illustrates that with the increasing nanoparticle size, the heat transfer is deteriorated at first, but then enhanced after the nanoparticle size exceeds the average surface roughness (100 nm). Narayan et al. (2007) and Das et al. (2008) suggested that when the nanoparticles size equals roughly to the surface roughness, the deposited nanoparticles could settle in the cavity and thus significantly reduce the active site density. On the contrary, when the nanoparticles are obviously larger or smaller than the surface roughness, the deposited nanoparticles could create more active sites and thus enhance the heat transfer. This hypothesis was verified in
this study, as shown in Figure 3.20(b). However, the increased nanoparticle size significantly reduces the intensity of Brownian motion, which leads to a decreased equivalent thermal conductivity $\lambda_{bm}$ and the proportion of $q_{bm}$, as shown in Figure 3.20(c) and (d), respectively.

Lastly, the effects of nanoparticle material were analyzed. Although a large number of materials could be used to prepare nanofluids, oxides, thanks to their physically and chemically stable properties, are widely regarded as promising materials for practical applications (Buongiorno et al., 2008). Therefore, several widely used oxides (SiO$_2$, Al$_2$O$_3$, TiO$_2$, ZnO and ZrO$_2$) as summarized by Vafaei and Borca-Tasciuc (2013) were selected in this study and some of their property parameters are listed in Table 3.3.

Table 3. 3 Physical Properties of the nanoparticle materials and water (webbook.nist.gov).

<table>
<thead>
<tr>
<th>Material</th>
<th>Density (kg/m$^3$)</th>
<th>Thermal conductivity (W/(m K))</th>
<th>Specific heat (J/(kg K))</th>
<th>$\gamma$ ( - )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Saturated water</td>
<td>958</td>
<td>0.679</td>
<td>4216</td>
<td>1.000</td>
</tr>
<tr>
<td>SiO$_2$</td>
<td>2410</td>
<td>1.4</td>
<td>705</td>
<td>0.931</td>
</tr>
<tr>
<td>Al$_2$O$_3$</td>
<td>3490</td>
<td>25.0</td>
<td>451</td>
<td>3.788</td>
</tr>
<tr>
<td>TiO$_2$</td>
<td>4010</td>
<td>8.3</td>
<td>690</td>
<td>2.894</td>
</tr>
<tr>
<td>ZnO</td>
<td>5606</td>
<td>3.2</td>
<td>580</td>
<td>1.948</td>
</tr>
<tr>
<td>ZrO$_2$</td>
<td>5570</td>
<td>2.2</td>
<td>480</td>
<td>1.465</td>
</tr>
</tbody>
</table>

Figure 3. 21: Effects of the nanoparticle material. (Note: 0.1 vol% nanofluids, nanoparticle size 34 nm, surface roughness 100 nm)
The numerical results are shown in Figure 3.21, which indicate that the heat transfer performance is strongly affected by the nanoparticle material. The highest heat transfer coefficient was predicted with the SiO$_2$/water nanofluid while the lowest heat transfer coefficient appeared with the Al$_2$O$_3$/water nanofluid (Figure 3.21(a)), despite SiO$_2$ has the lowest thermal conductivity while Al$_2$O$_3$ has the highest thermal conductivity among the selected materials (Table 3.3). However, no explicit impact of the nanoparticle material was predicted on the equivalent thermal conductivity $\lambda_{bm}$. Meanwhile, the effects of nanoparticle material on the active site density and the Brownian motion component $q_{bm}$ was not clearly presented.

![Figure 3.22: Effects of the nanoparticle material on the quenching and evaporation heat flux components.](image)

In order to achieve a deeper insight, the effects of nanoparticle materials on the quenching and evaporation heat flux components were further analyzed, as shown in Figure 3.22. The results demonstrate that the significances of the quenching and evaporation mechanisms – both correlated to the active site density – decrease with the increased wall-liquid interaction parameter $\gamma$, which actually improves the significance of the convection and Brownian motion mechanisms. Considering the negligibly changed $\lambda_{bm}$ (Figure 3.21(c)), it could be suggested that the effects of nanoparticle materials on nucleate boiling of nanofluids are implemented mainly through altering the characteristics of bubble nucleation, which indirectly change the importance of the $q_{bm}$ component through elevating or reducing the temperature difference for heat
transfer. Comparatively, the change in nanoparticle material alone does not significantly change the intensity of nanoparticle Brownian motion and its equivalent thermal conductivity.

3.2.4 Conclusions

In this study, a new HFP model was proposed for nucleate boiling of nanofluids. Compared with the classic HFP model, the new model contains an additional HFP component that accounts for the heat transfer by the nanoparticle Brownian motion in the microlayer. Numerical computations were conducted using both the new and classic HFP models. The numerical results were analyzed and compared against the experimental data available in the literature. The conclusions arising from this study are as follows:

(1) Due to the continuously increased nanoparticle concentration in the microlayer, heat transfer by the Brownian motion of nanoparticles in the microlayer becomes an important mechanism of heat removal from the heater surfaces boiling in nanofluids.

(2) The new HFP model achieves a better agreement with the experimental data than the classic HFP model, especially when the applied heat flux is high. This indicates that the active site density available on the heater surface plays a crucial role in determining the significance of nanoparticle Brownian motion.

(3) For dilute nanofluids, the heat transfer due to nanoparticle Brownian motion is positively affected by the bulk concentration and negatively influenced by the nanoparticle size. An increased bulk concentration or a decreased nanoparticle size would enhance the significance of nanoparticle Brownian motion in heat removal. Comparatively, the nanoparticle material does not have much impact on the heat transfer due to the nanoparticle Brownian motion.
Chapter 4

Numerical Modelling of Two-phase Flows of Dilute Nanofluids

The main findings of this chapter have been included in:

4.1 Two-fluid Modelling of Air-nanofluid Bubbly Flows

Abstract:

The bubbly flows of air-nanofluid and air-water in a vertical tube were numerically simulated using the two-fluid model. Comparison of the numerical results against the experimental data of Park and Chang (2011) demonstrated that the classic two-fluid model, although agreed well with the air-water data, was not applicable to the air-nanofluid bubbly flow. It was suggested that in a bubbly flow system, the existence of interfaces allows the spontaneous formation of a thin layer of nanoparticle assembly at the interfaces, which significantly changes the interfacial behaviours of the air bubbles and the roles of the interfacial forces. As the conservation equations of the classic two-fluid model are still applicable to nanofluids, the mechanisms underlying the modified interfacial behaviours need to be carefully taken into account when modelling air-nanofluid bubbly flows. Thus, one of the key tasks when modelling bubbly flows of air-nanofluid using the two-fluid model is to reformulate the interfacial transfer terms according to the interfacial behaviour modifications induced by nanoparticles.

4.1.1 Introduction

As a new type of engineered liquids for enhancing heat transfer, nanofluids have been attracting an increasing attention since the novel concept “nanofluid” was firstly proposed by Choi and Eastman (1995). Nanofluids were initially investigated because of their improved thermal conductivity brought out by the nanoparticles. During the past years, numerous studies have been conducted on the convective transport phenomena in nanofluids (Buongiorno, 2006). Up to today, agreements have been reached on the mechanisms of heat transfer in single-phase nanofluids (Chandrasekar et al., 2012; Yu et al., 2012). It is generally accepted that due to their small sizes, nanoparticles are mixed with the base liquid at near-molecular level. A dilute nanofluid behaves hydro-dynamically like its pure base liquid and could be treated theoretically as a single-phase liquid. This has allowed developing predictive models for single-phase flows of nanofluids based on the Navier-Stokes equations (Kamyar et al., 2012).
Existing studies (Akbari et al., 2011; Moraveji and Ardehali, 2013) have proven that the single-phase computational fluid dynamics (CFD) model is capable of describing the flow and heat transfer behaviours in nanofluids on condition that the thermodynamic properties are properly formulated.

In recent years, the great potential of enhancing heat transfer using two-phase flows of nanofluids, especially by nucleate boiling, has been gradually recognized (Cheng et al., 2008). However, due to the relative novelty and inherent complexity, agreements are far to be reached in this area and many opinions are still in controversy (Barber et al., 2011). Nanofluids come with various concentrations, however, dilute nanofluids with very low nanoparticle loads (typically less than 0.1 vol%) are generally preferred for boiling applications (Buongiorno et al., 2009) when one considers the practical feasibility. For nanofluids with such low concentrations, a number of experimental measurements demonstrated that their physical properties (e.g. the thermal conductivity, density, viscosity, specific heat and latent heat) are negligibly different from those of their pure base liquids (Kim et al., 2007; Kwark, 2009). The dramatically changed boiling heat transfer performances have been attributed to the surface modifications induced by nanoparticle deposition during the boiling process (Wen et al., 2011; Vafaei and Borca-Tasciuc, 2014). In recent years, CFD modellings of nucleate boiling of nanofluids have been conducted (Li et al., 2014a; Li et al., 2014b; Li et al., 2015) based on the two-fluid model of Ishii (Ishii, 1975). In these studies, the effects of nanoparticle deposition on bubble nucleation on the heater surface were properly considered. The model applicability and accuracy, although still not satisfactory, have been largely improved. However, an important fact may have been ignored – the nanoparticles suspended in the base liquid not only modify the heat surface, but also change the two-phase flow structures and hydrodynamic features.

Nayak et al. (2011) studied experimentally the transient and stability behaviours of boiling two-phase natural circulation loop with water and Al₂O₃/water nanofluid (1.0 wt%, approx. 0.25 vol%), respectively. They found that the natural circulation flow behaviours of nanofluid were very close to that of water in single-phase conditions. However, the buoyancy induced flow rates in boiling conditions were relatively higher with nanofluid than with water. Dominguez-Ontiveros et al. (2010) observed the pool
boiling of Al$_2$O$_3$/water nanofluids (0.001 and 0.002 vol%) using dynamic particle image velocimetry (DPIV). They found that the hydrodynamic behaviours of bubbles were significantly changed when nanoparticles are introduced into water. Recently, Rana et al. (2014) measured the void fraction in boiling flows of ZnO/water nanofluids (0.001–0.01 vol%). The results revealed that the void fraction decreased down to 86% with the use of nanofluid in place of water.

In addition, the modifications of two-phase flow characteristics by nanoparticles were also observed in isothermal flows. Wang and Bao (2009) investigated the transition of two-phase flow regimes in a vertical capillary tube, using nitrogen as the gaseous phase and CuO/water nanofluid (0.5 wt%, approx. 0.08 vol%) and pure water as the liquid phase, respectively. They found that the bubbly-slug flow regime transition occurred at a lower liquid superficial velocity or a higher gas superficial velocity in the nanofluid than in water. This indicated that nanofluids could maintain a bubbly flow pattern with a higher void fraction than pure water, which is undoubtedly of great importance to enhancing two-phase heat and mass transfers, thanks to the larger interfacial area created by the higher void fraction in nanofluids. Wang and Bao (2009) suggested that the changed flow-regime transition characteristics were mainly due to the changed liquid surface tension. Park and Chang (2011) measured the local distributions of air-liquid bubbly flow parameters in a vertical tube using a conductivity double-sensor probe. Both pure water and Al$_2$O$_3$/water nanofluid (0.1 vol%) were used as the working liquids. The results showed that when the operational conditions were exactly the same, the air-nanofluid bubbly flow had a more flattened void fraction distribution, lower bubble velocity, higher interfacial area concentration and small bubble size than those in the air-water flow. They attributed these changes to the altered interfacial drag and lift forces.

Although the physical mechanisms underlying the flow modifications are yet to be discovered, it is evident that the existence of nanoparticles in the liquid has a significant effect on the two-phase flow structures and features, even with extremely low nanoparticle concentrations. As two-phase flows are coupled systems, an effective CFD simulation of two-phase flows requires accurate description of the inter-phase transport processes of mass, momentum and energy in the whole flow field. Therefore, in order
to achieve an effective modelling of two-phase flows of nanofluids using the two-fluid model, the closure correlations, which are generally empirical or semi-empirical and thus not universal, have to be carefully reformulated or selected in order to account for the specific features induced by nanoparticles.

In order to identify the individual factors affecting the hydrodynamic behaviours of nanofluid two-phase flows, isothermal bubbly flow of air-nanofluid in a vertical tube was modelled in this study using the classic two-fluid model incorporated with various inter-phase transfer terms. Two-phase flow parameters including the air velocity and void fraction were predicted and compared against the experimental data of Park and Chang (2011). Bubbly flow of air-water was also simulated for the purpose of comparison. The results demonstrated that the classic two-fluid model had a satisfactory accuracy for the air-water bubbly flow, but was inapplicable to the air-nanofluid flow. Further analyses demonstrated that the suspended nanoparticles in the liquid tend spontaneously to assembly at the interfaces, which significantly changes the liquid-bubble interfacial behaviours and makes the existing empirical closure correlation invalid to the air bubbles submerged in nanofluids. Suggestions were given for future studies.

4.1.2 Modelling of Bubbly Flow in a Vertical Tube

4.1.2.1 The Two-fluid Model

The experimental data of Park and Chang (2011) were employed in this study for model validation and comparison. In their experiments, dilute Al₂O₃/water nanofluid with a concentration of 0.1 vol% was synthesized by dispersing γ-Al₂O₃ nanoparticles (mean diameter 25 nm) into distilled water. Then, the nanofluid was supplied into a vertical acrylic tube (15 mm in diameter and 2.5 m in height) from the bottom. Air bubbles were also generated at the bottom using a bubble bed. The mixture was driven by a pump to form an upward two-phase flow in the test section. The experiments were conducted under the atmospheric pressure and the ambient temperature. By controlling the superficial velocities at \( j_l = 2.83 \text{ m/s} \) for the liquid and \( j_a = 0.19 \text{ m/s} \) for the air, respectively, a stable bubbly flow was achieved in the tube. Radial distributions of the two-phase flow parameters including the void fraction, bubble velocity and diameter
were measured using a conductivity double-sensor probe at a height of 1.75 m downstream of the tube inlet, which was far enough for a fully developed flow. Experiments were also conducted using pure water in place of the nanofluid.

Based on the experimental conditions, the two-fluid model (Ishii, 1975) was selected to model the flow. As the flows were isothermal, the energy equation and the interphase mass transfer terms were excluded from the model. Thus, the conservation equations take the following forms:

The continuity equation

$$\frac{\partial}{\partial t} \left( \alpha_k \rho_k \right) + \nabla \cdot \left( \alpha_k \rho_k \bar{U}_k \right) = 0$$  \hspace{1cm} (4.1)

The momentum equation

$$\frac{\partial}{\partial t} \left( \alpha_k \rho_k \bar{U}_k \right) + \nabla \cdot \left( \alpha_k \left( \rho_k \bar{U}_k \bar{U}_k - \mu_k \left( \nabla \bar{U}_k + \left( \nabla \bar{U}_k \right)^T \right) \right) \right)
= \alpha_k \left( S_{\text{bious}} - \nabla P_k \right) + \bar{F}_k$$  \hspace{1cm} (4.2)

where, $k$ is the phase denotation ($k = g$ for the gaseous phase and $k = l$ for the liquid phase). $\alpha$, $\rho$, $U$ and $P$ represent the volume fraction, density, velocity and pressure, respectively. $\bar{F}_i$ represents the interfacial forces, including the drag force $\bar{F}_D$, lift force $\bar{F}_L$, turbulent dispersion force $\bar{F}_{TD}$ and wall lubrication force $\bar{F}_W$, respectively.

$$\bar{F}_k = \bar{F}_D + \bar{F}_L + \bar{F}_{TD} + \bar{F}_W$$  \hspace{1cm} (4.3)

When the spherical-bubble assumption (Ansys, 2011) was employed in this study, the forces were defined by

$$\bar{F}_D = \frac{3}{4} \frac{C_D}{d_b} \alpha_g \rho_l \left| \bar{U}_g - \bar{U}_l \right| \left( \bar{U}_g - \bar{U}_l \right) \hspace{1cm} (4.4)$$

$$\bar{F}_L = C_L \alpha_g \rho_l \left( \bar{U}_g - \bar{U}_l \right) \times \left( \nabla \times \bar{U}_l \right) \hspace{1cm} (4.5)$$

$$\bar{F}_{TD} = -C_{TD} \rho_l \kappa \alpha_l \hspace{1cm} (4.6)$$

$$\bar{F}_W = -\frac{\alpha_g \rho_l \left( \bar{U}_g - \bar{U}_l \right)}{d_b} \max \left( 0, C_{W1} + C_{W2} \frac{d_b}{y_w} \right) \bar{n}$$  \hspace{1cm} (4.7)

where, $C_D$, $C_L$, $C_{TD}$, $C_{W1}$ and $C_{W2}$ are empirical coefficients which need to be carefully determined. Formulation of these coefficients is one of the most critical tasks when modelling bubbly flows using the two-fluid model. During the past decades, a number
of empirical or semi-empirical correlations, including the Ishii-Zuber (Ishii and Zuber, 1979) and Grace (Clift et al., 1978) models for \( C_D \) and the Tomiyama correlation for \( C_L \) (Tomiyama, 1998), have been proposed. However, the applicability of these correlations to bubbly flow of nanofluids is still open to question, as will be discussed in the following sections.

4.1.2.2 Numerical Procedures

Due to the axis-symmetric distribution of the two-phase flow field in the tube, a quarter of the test section (Park and Chang, 2011) was built as the computational domain, as illustrated in Figure 4.1. The domain was then discretized using structured hexahedral meshes with finer mesh close to the tube wall and coarser mesh at the tube centre. The centre-to-wall mesh size ratio was 2.5. Uniform mesh size (4 mm) was employed in the axial direction of the tube. Mesh sensitivity test proved that mesh independence was achieved at 63,000 cells since a further increase of the cell number to 128,000 just caused a small change less than 1% in the predicted air velocity at a randomly selected monitoring point.

During the computations, uniformly distributed air and liquid flow rates were applied at the inlet and a zero pressure boundary condition was applied at the outlet. The flow of the gaseous phase was assumed to be laminar and turbulence was only modelled for the liquid phase using the updated \( k-\epsilon \) model by Sato and Sekoguchi (1975), where a bubble-induced additional turbulent viscosity was considered when estimating the liquid effective viscosity \( \mu^e_L \):

\[
\mu^e_L = \mu_l + \rho_l C_{\mu} \frac{k_l^2}{\epsilon_l} + C_{\mu b} d_b \alpha_s \left| U_s - \overline{U}_l \right| \tag{4.8}
\]

The items on the right-hand-side of Equation 4.8 represent the molecular viscosity, turbulent viscosity and bubble-induced additional turbulent viscosity, respectively.

The aforementioned model equations were solved using the commercial CFD code ANSYS-CFX 14.5. Convergence was achieved within 2,000 iterations when the residual of the liquid continuity equation dropped down to lower than \( 1 \times 10^{-5} \).
4.1.3 Results and Discussion

4.1.3.1 Model Applicability to Water and Nanofluid

Air-water bubbly flow was firstly computed using the two-fluid model. Bubble coalescence and breakup were not considered. Instead, a uniform bubble diameter of $d_b = 6$ mm was estimated based on the experimental data (Park and Chang, 2011). For the calculation of interfacial forces, constants were selected for the turbulent dispersion coefficient ($C_{TD} = 0.1$) and wall lubrication coefficients ($C_{W1} = -0.01$ and $C_{W2} = 0.05$), as recommended by CFX-14.5. The drag coefficient was calculated using the Ishii-Zuber model (Ishii and Zuber, 1979) which is a function of the bubble Reynolds number

$$C_D = \frac{24}{Re_b} \left( 1 + 0.1Re_b^{0.75} \right) \quad 0 \leq Re_b \leq 500 \sim 1000$$

The lift coefficient was calculated using the Tomiyama correlation (Tomiyama, 1998) in order to account for the variable acting direction of the lift force depending on the bubble size.
where, \( f(Eo^*) \) is an empirical correlation of the modified Eötvös number (Tomiyama, 1998).

\[
f(Eo^*) = 0.00105Eo^*^3 - 0.0159Eo^*^2 - 0.0204Eo^* + 0.474 \tag{4.11}
\]

\[
Eo^* = \frac{g \left( \rho_l - \rho_g \right) d_h^2}{\sigma} \tag{4.12}
\]

where, \( d_h \) is the maximum bubble dimension in the flow direction and was estimated using the Wellek correlation (Wellek et al., 1966).

The two-fluid model incorporated with the above closure correlations is termed in this study as the classic two-fluid model (TFM). The predicted radial distributions of void fraction and bubble velocity at \( Z = 1.75 \) m are shown in Figure 4.2. The comparison demonstrated that the predicted local two-phase flow parameters agreed well with the experimental data (Park and Chang, 2011), which proved the validity of the classic two-fluid model to air-water bubbly flows.
Figure 4. 2: Comparison the classic two-fluid model against the experimental data of water: (a) void fraction; (b) bubble velocity (Park and Chang, 2011).

Then, the air-nanofluid bubbly flow (Park and Chang, 2011) was computed using the classic two fluid model with the same correlations (Equation 4.9 and 4.10) for $C_D$ and $C_L$. For the air-nanofluid case, the average bubble size was estimated to $d_b = 3$ mm according to the experimental data (Park and Chang, 2011). The liquid density and viscosity were calculated using Equation 4.13 and 4.14 (Prasher et al., 2006), respectively.

$$\rho_{nf} = \rho_w (1 - \phi_v) + \rho_{np} \phi_v$$  \hspace{1cm} (4.13)

$$\mu_{nf} = (1 + 2.5 \phi_v) \mu_w$$  \hspace{1cm} (4.14)

where, $\phi_v$ is the volumetric concentration of nanoparticles in the nanofluid, $\rho_{np}$ stands for the nanoparticle density. For the dilute nanofluid of this study ($\phi_v = 0.1$ vol%), the effects of nanoparticle on its density and viscosity could be safely ignored, which is consistent with most experimental measurements (Kim et al., 2007; Kwark, 2009). However, the existence of nanoparticles in water was found to reduce the liquid surface tension to a measurable extent. Esmaeilzadeh et al. (2014) used a series of advanced techniques including dynamic light scattering, zeta potential measurement and centrifugation to study the effects of nanoparticles on the air-water surface tension. They found that the addition of ZrO$_2$ nanoparticles into water could alter the liquid surface activity and reduce the surface tension. The experiments by Kwark (2009)
further revealed that even the addition of an extremely small amount of Al$_2$O$_3$ nanoparticles (0.001 g/l, approx. 2.5×10$^{-6}$ vol%) could cause a 2 % reduction in the liquid surface tension. When the CuO nanoparticle concentration in pure water increased up to 1.0 wt% (approx. 0.16 vol%), the liquid surface tension reduction could be as large as 15% (Wang and Bao, 2009). For the 0.1 vol% Al$_2$O$_3$/water nanofluid of Park and Chang (2011), the liquid surface tension was estimated based on Kwark’s measurements (2009) to be 95% of that of pure water.

Figure 4. 3: Comparison the classic two-fluid model against the experimental data of nanofluid: (a) void fraction; (b) bubble velocity (Park and Chang, 2011).
Figure 4.4: Prediction of the void fraction development along the tube using the TFM. Note: Due to the large length-to-diameter ratio of the computational domain, the void fraction contours were not shown in actual proportion.

The predicted local bubbly flow parameters of air-nanofluid are shown in Figure 4.3. The two-fluid model predicted a near-wall-peaked distribution of the void fraction (Figure 4.3(a)), which was totally different from the actual central-peaked distribution as observed by Park and Chang (2011), despite the predicted bubble velocity was just slightly larger than the experimental data (Figure 4.3(b)).

The computations using the classic two-fluid model returned totally different bubble distributions in the tube. In order achieve a clear view of the bubble migration prediction, the simulated void fraction profiles in a section plan along the tube axis were shown in Figure 4.4, for air-water and air-nanofluid bubbly flows, respectively. Figure 4.4 illustrates that for both air-water and air-nanofluid bubbly flows, the two-phase flows only need a short distance to reach full-development. In the air-water bubbly flow case, when bubbles were assumed to be uniformly injected from the bottom of the tube, they quickly moved towards the tube centre. On the contrary, when the bubbles are injected to nanofluid, they were falsely predicted to move towards the
tube wall when moving downwards with the liquid. Therefore, it’s evident that the classic two-fluid model, although has been widely validated to be effective for bubbly flows of water, is not applicable to that of nanofluids despite the negligibly changed liquid properties. In order to achieve an effective modelling of bubbly flow of nanofluids, the two-fluid model has to be carefully modified.

4.1.3.2 Model Improvement for Air-nanofluid Bubbly Flows

As shown in Figure 4.2 and 4.3, the void fraction was flattened with smaller bubbles in the air-nanofluid case than in the air-water case. Park and Chang (2011) proposed that among the interphase forces ($\vec{F}_D$, $\vec{F}_L$, $\vec{F}_{TD}$ and $\vec{F}_W$), the determinant of the transverse motion of bubbles is the interaction between the drag force and the lift force. They evaluated the effects of the drag coefficient based on the experimentally measured bubble size, using the Grace model (Grace and Weber, 1982).

$$C_D = \frac{4}{3} \frac{g d_b}{u_f^2} \frac{\rho_l - \rho_g}{\rho_l}$$

(4.15)

They found that the drag coefficient in the nanofluid is around 6% larger than that in water with the same bubble size. It was noticed that the Grace model (Equation 4.15) is appropriate for sparsely dispersed fluid particles. Considering the dense bubble effects in some local region as the void fraction near the tube axis approached 0.2 (Figure 4.3(a)), the drag force was further evaluated in this study by using the Ishii-Zuber model (Ishii and Zuber, 1979) (Equation 4.9). The results demonstrated that for the flow conditions of this study, the Ishii-Zuber model and the Grace model generated very close predictions, as shown in Figure 4.5. In addition, the drag coefficients as a function of the bubble size for the air-water and air-nanofluid cases generated from the Ishii-Zuber model were very close, as illustrated in Figure 4.6. Therefore, the variation in the drag force induced by the nanoparticles was not expected to be responsible for the significant deviation as observed in Figure 4.3.
Figure 4.5: Comparison of the Ishii-Zuber model (Ishii and Zuber, 1979) and Grace model (Grace and Weber, 1982) for drag force modelling.

Figure 4.6: The drag coefficient calculated by the Ishii-Zuber model (Ishii and Zuber, 1979).

The lift force acts in the directions perpendicular to the flow. According to Tomiyama (1998), the lift force would change its sign with increasing bubble size, which causes larger bubbles move transversely towards the axis while smaller bubbles move towards the wall. The lift coefficient as calculated by the Tomiyama correlation (Equation 4.10) is plotted in terms of the bubble size in Figure 4.7, for bubbles in water.
and nanofluid, respectively. For the purpose of comparison, the Hibiki-Ishii correlation (Hibiki and Ishii, 2007) (Equation 4.16), which also yields a negative lift coefficient for larger bubbles, was plotted as well.

\[
C_L = \xi \left( (C_L^{\text{low}}(R_{eb}, G_s))^2 + (C_L^{\text{high}}(R_{eb}))^2 \right) \tag{4.16}
\]

where, \(C_L^{\text{low}}\) and \(C_L^{\text{high}}\) are empirical piecewise functions of the bubble Reynolds number \(R_{eb}\) and the non-dimensional shear rate \(G_s\).

\[
G_s = \frac{d_b}{2(U_e - U_i)} \left| \frac{d \bar{U}_z}{dx} \right| \tag{4.17}
\]

Figure 4.7 illustrates that for the air-water case, a negative lift coefficient was yielded from Equation 4.10 at \(d_b = 6\) mm, which caused the lift force pointing towards the axis. This agreed well with the experimental observations of the air-water bubbly flow (Figure 4.2(a)) (Park and Chang, 2011). For the air-nanofluid case, however, the Tomiyama correlation generated a positive lift coefficient (\(C_L = 0.288\)) at \(d_b = 3\) mm, causing the lift force pointing towards the wall and a near-wall peaked void fraction distribution was predicted (Figure 4.3(a)). For air-water bubbly flow, the Hibiki-Ishii correlation achieved a very close prediction to that of the Tomiyama correlation.

![Figure 4.7: The lift coefficient changes as a function of bubble size.](image)
As the actual void fraction distribution was central-peaked in the air-nanofluid bubbly flow with \(d_b = 3\) mm (Figure 4.3(a)), it was expected that with the increasing bubble size, the positive-to-negative transition in the lift coefficient appears at a smaller bubble size in nanofluid than in water. Unfortunately, due to the insufficient fundamental investigations on this issue, a quantitative correlation for estimating \(C_L\) in nanofluids is still absent. For the air-nanofluid bubbly flow of this study, a satisfactory agreement was achieved between the numerical results and the experimental data when
the lift coefficient took the value \( C_L = -0.03 \), as shown in Figure 4.8(a). A larger negative lift coefficient led to a higher central peak of the void fraction distribution. When a positive lift coefficient was applied, the peak of void fraction gradually moved towards the wall. Therefore, the modelling of lift force has a significant effect on the predicted distribution of void fraction. However, the air bubble velocity distribution seemed to be insensitive to the lift coefficient (Figure 4.8(b)).

The calculation of lift force has long been a challenging task when modelling bubbly flows. Hibiki and Ishii (2007) conducted a comprehensive survey of the lift force correlations available in the literature. According to the survey, most lift force correlations are empirical or, at least, semi-empirical. The existence of nanoparticles in the liquid further intensifies the complexity and further fundamental studies are in urgent demand in this area.

Currently, the transport and thermodynamic properties of nanofluids were mostly measured and characterized under static conditions. However, according to the study by Vermant and Solomon (2005), the application of flow could cause various novel microstructure states in colloid suspension, which are strongly affected by the balance among inter-particle forces, Brownian motion and hydrodynamic interactions. The resulting nonequilibrium microstructure is a principal determinant of the suspension rheology and the force balance on bubbles. This was perhaps the major reason responsible for the earlier-appearing positive-to-negative transition of the lift coefficient. Unfortunately, the effects of nanoparticles on the inter-phase forces have been rarely investigated and the mechanisms are still unknown.

### 4.1.3.3 Effects of Nanoparticles on the Interfacial Behaviours

As observed by Park and Chang (2011), one of the most distinct characteristic of the air-nanofluid flow, when compared with the air-water flow, was the smaller bubble size. For a spherical bubble submerged in quiescent liquid, its equilibrium size could be estimated by the Young-Laplace equation

\[
\Delta P = \frac{2\sigma}{d_b/2}
\]

(4.18)
where, $\Delta P$ is the pressure difference between in and out of the bubble. Equation 4.18 indicates that the decreased surface tension would lead to a smaller bubble size in order to maintain the force balance. However, according to the experimental measurements by Kwark (2009), the addition of 0.1 vol% $\text{Al}_2\text{O}_3$ nanoparticles into pure water just caused a 5% reduction in the liquid surface tension, which was not expected to be fully responsible for the significant bubble diameter decrease from 6 mm to 3 mm. Thus, there should be some other factors impacting the interfacial behaviours of the air-nanofluid flow.

![Image](image_url)

Figure 4.9: Fluorescence confocal microscope image of water droplets dispersed in toluence, covered with CdSe nanoparticles (Lin et al., 2005).

Thermodynamically, all systems have the tendency to minimize their energy spontaneously in order to reach a stable condition. Due to the high interfacial energy induced by the small particle size, colloid suspensions of nanoparticles in liquid are thermodynamically unstable. Thus, the phenomenon of nanoparticle clustering in the liquid (Zhou et al., 2014) and self-assembly at the fluid interface (Lin et al., 2005; Blute et al., 2009) has been widely observed, which was believed to be driven by the minimization of the Helmholtz free energy (Lin et al., 2005; Popp et al., 2010). Shown in Figure 4.9 is a confocal microscope image of the self-assembly of fluorescent nanoparticles (CdSe) at water-toluence interface, as observed by Lin et al. (2005). Figure 4.9 demonstrates that a thin layer of nanoparticles were absorbed at the interface. Kim et al. (2014) reported that this thin layer of nanoparticles could enhance mass transfer between the phases by thinning the diffusion boundary layer around the
bubbles. Grzelczak et al. (2010) further pointed out that the macroscopic viscous flow could enhance nanoparticle self-assembly at the interfaces, this is especially true for particles from 100 nm to 1 μm in diameter. They suggested that the shear rate and shear strain, nanoparticle volume fraction, particle interaction potentials and poly-dispersity are the key factors that affect flow-induced nanoparticle self-assembly (Grzelczak et al., 2010).

The absorbed nanoparticle assembly at the interface was found to be able to stabilize the bubbles mechanistically and could effectively impede smaller bubbles coalescing into larger bubbles. This was perhaps the reason responsible for the smaller air bubble size in the Al₂O₃/water nanofluid than in pure water as observed by Park and Chang (2011), as well as the bubbly flow with higher void fraction in CuO/water nanofluid than in pure water as observed by Wang and Bao (2009). The stabilizing function of nanoparticles on gas bubbles in liquid has been widely recognized and utilized to fabricate liquid foams with fine textures (Worthen et al., 2013) and capsule shells for high-efficiency drug delivery (Ariga et al., 2011). Unfortunately, the effects of nanoparticle self-assembly on the interfacial behaviours in bubbly flows have not been fully investigated. Substantial fundamental studies should be conducted on this regard in the future.

4.1.4 Conclusions

Bubbly flows of air-water and air-nanofluid were numerically investigated using the two-fluid model. Comparison of the numerical results against the experimental data available in the literature revealed that the classic two-fluid model agreed well with the experimental data of air-water bubbly flows, but needed substantial improvement in order to achieve an effective modelling of air-nanofluid bubbly flows. The effects of nanoparticles on the interfacial behaviours and interphase transport mechanisms were analysed based on the experimental observations in the literature. Conclusions arising from this study are as follows:

1. Although the addition of a small amount of nanoparticles into the base liquid does not cause measurable changes in the liquid properties, the spontaneous nanoparticle self-assembly at the interface could significantly change the
interfacial behaviours of the air bubbles. This was supposed to be the major reason responsible for the distinctly changed two-phase flow characteristics (e.g., smaller bubble size) of air-nanofluid bubbly flows than those of air-water flows.

(2) As the governing equations are still applicable to nanofluids, the key job when modelling air-nanofluid bubbly flows using the two-fluid model is to formulate the interphase transport terms in order to take into account the specific features induced by the existence of particles. This study demonstrates that the lift force has different acting roles in nanofluid than in pure water, which causes the lift force reverses its direction at a smaller bubble size.
4.2 MUlItiple-SIze-Group (MUSIG) Modelling of Air-nanofluid Bubbly Flows in a Vertical Tube

Abstract

The MUtiple-SIze-Group (MUSIG) model was used in this study to simulate bubbly flows of air-water and air-nanofluid in a vertical tube. Flow parameters including the void fraction, gas velocity, interfacial area concentration and Sauter mean bubble diameter were predicted and compared against the experimental data available in the literature. The model agreed well with the experimental data of air-water bubbly flow, but exhibited notable discrepancies from the data of air-nanofluid bubbly flow. With the aim to improve the MUSIG model for an effective modelling of air-nanofluid bubbly flows, some latest experimental and theoretical research outcomes were summarized and analysed. It was proposed that the key job when modelling bubbly flows of nanofluids using the MUSIG model is to address the spontaneous assembly of nanoparticles at bubble surfaces and its effects on the interfacial forces and bubble coalescence process.

4.2.1 Introduction

Over the pass decades, great efforts have been devoted to the development of advanced fluids offering better heat transfer performances for a variety of thermal management systems. Among them, “nanofluid” which was proposed by Choi (1995) is regarded promising. Nanofluids are a new type of engineered fluids that consist of uniformly dispersed nanometre-sized particles in common base liquids. Novel features of nanofluids, such as enhanced thermophysical properties (Khanafer and Vafai, 2011), single-phase convective (Kakaç and Pramuanjaroenkij, 2009) and boiling heat transfer performances (Jacqueline et al., 2011), have been reported numerously. It has been widely accepted that the addition of nanoparticles can significantly increase the forced convective and boiling heat transfer of base liquids (Jo et al., 2009; Kim, 2009; Rana et al., 2013). The existing studies mostly focused on the heat transfer characteristics of nanofluids, while less attention has been paid to their basic hydraulic phenomena. In fact, the heat transfer performance of nanofluids, particularly their two-phase flows, is
closely related to their flow structures (Atmane and Murray, 2005), since a large proportion of the heat and mass are transferred through the liquid-bubble surfaces. An in-depth understanding of the hydrodynamic behaviours of nanofluids is critical to the further extension of their heat transfer applications.

Significant impact of the existence of nanoparticles on two-phase flow structures has been revealed by numerous studies. Using the high-speed visualization and image processing technique, Rana et al. (2014) measured the void fraction in the flow boiling of water-ZnO nanofluids (0.001~0.01 vol%) and detected a significant void fraction decrease (up to 86%) compared to pure water. Dominguez-Ontiveros et al. (2010) measured the phase velocities in a boiling pool using the dynamic particle image velocimetry (DPIV), and found that in Al2O3/water nanofluids (0.001 and 0.002 vol%) the fluid velocities were generally depressed relative to the pure water case. Recently, observations of modified two-phase flow regimes in isothermal flows were also reported by Wang and Bao (2009) who investigated the two-phase flow patterns of nitrogen bubbles in CuO/water nanofluids (0.5 wt%, approx. 0.08 vol%) in a vertical capillary tube. They found that the bubbly-to-slug flow pattern transition in nitrogen-nanofluid flows occurred at a lower liquid velocity compared to nitrogen-water flows, which indicated that the nitrogen-nanofluid flow could stay bubbly with a higher void fraction.

Bubbly flows are generally multi-dispersed systems where the bubbles have a large spectrum of sizes and shapes in the liquid. How to model the dynamic evolution of these dispersed bubbles has been the key concern of two-phase flow simulations. When investigating such cases, the MUltiple-SIze-Group (MUSIG) model (Lo, 1996) which provides an efficient method for solving the population balance theory within the classic Eulerian-Eulerian framework has been widely employed. Alongside the conservation equations and population balance equations, a number of closure equations for interfacial transport of mass, momentum and energy as well as bubble coalescence and break-up are incorporated in the MUSIG model. Appropriate formulation of these closure equations is the key determination of the overall predictive accuracy. Although a number of closure equations, including the Ishii-Zuber drag model (Ishii and Zuber, 1979) and the Tomiyama lift model (Tomiyama, 1998), have
been fully validated for bubbly flows of pure liquids with or without heat transfer (Li et al., 2006; Cheung et al., 2007), these closure equations are still the weakest link due to their empirical nature (Ishii and Mishima, 1984). When modelling bubbly flows of nanofluids, there naturally rises the question whether they are still applicable, in view of the modified properties and two-phase flow structures.

In order to develop a predictive model for multi-dispersed bubbly flows of nanofluids, the MUSIG model was employed in this study as a theoretical frame to model the bubbly flows of air-nanofluid. Air-water bubbly flow was also simulated for the purpose of comparison. Two-phase flow parameters including void fraction, gas velocity, interfacial area concentration (IAC) and Sauter mean bubble diameter were predicted and compared against the experimental data of Park and Chang (2011). Through mechanistic analyses, the applicability of classic closure equations was examined and the impact of nanoparticles on the flow parameters was discussed. Finally, the numerical results were used to evaluate possible modifications to the existing closure equations with the aim to improve the MUSIG model for nanofluid bubbly flows.

### 4.2.2 The MUSIG Model

#### 4.2.2.1 The Flow Equations

Our previous studies (Li et al., 2015) have proven that the Eulerian-Eulerian framework is still applicable to air-nanofluid bubbly flows, given that nanoparticles and base fluid are mixed at a near-molecular level. For an isothermal bubbly flow without interphase mass and heat transfer, only mass and momentum conservations are considered. In the MUSIG model, the air bubbles are firstly assumed to be spherical and then divided into \(N\) size groups according to their diameter. Continuity equations of each size group are solved to capture the size distribution. Then the model is further simplified by assuming that all the bubbles are moving at the same velocity in a given control volume, so that only one set of momentum equations are solved for all bubble groups. Therefore, the MUSIG model of this study takes the following form:

The continuity equation of liquid phase
\[
\frac{\partial}{\partial t} (\alpha_i \rho_i) + \nabla \cdot (\alpha_i \rho_i \vec{U}_i) = 0 \quad (4.19)
\]

The continuity equation of gas phase (for the \(i\)th group bubbles, \(i=1 \sim N\))

\[
\frac{\partial}{\partial t} (\alpha_s \rho_s f_i) + \nabla \cdot (\alpha_s \rho_s \vec{U}_s f_i) = \left( \sum_{j=1}^{4} \Gamma_j \right) \quad (4.20)
\]

The momentum equation of liquid phase

\[
\frac{\partial}{\partial t} (\alpha_i \rho_i \vec{U}_i) + \nabla \cdot \left( \alpha_i \left( \rho_i \vec{U}_i \vec{U}_i - \mu_i \left( \nabla \vec{U}_i + (\nabla \vec{U}_i)^T \right) \right) \right) = \alpha_i (\rho_i g - \nabla P_i) + \vec{F}_{lg} \quad (4.21)
\]

The momentum equation of gas phase

\[
\frac{\partial}{\partial t} (\alpha_s \rho_s \vec{U}_s) + \nabla \cdot \left( \alpha_s \left( \rho_s \vec{U}_s \vec{U}_s - \mu_s \left( \nabla \vec{U}_s + (\nabla \vec{U}_s)^T \right) \right) \right) = \alpha_s \left( \rho_s g - \nabla P_{lg} \right) + \vec{F}_{gl} \quad (4.22)
\]

where the subscripts \(l\) and \(g\) are phase denotations (\(l\) for the liquid phase and \(g\) for the gas phase); \(\alpha\), \(\rho\), \(\vec{U}\), \(f_i\), and \(\vec{F}_{lg} (= - \vec{F}_{gl})\) represent the local void fraction, density, velocity, MUSIG volume fraction, and interfacial forces, respectively. The MUSIG volume fraction \(f_i\) is defined by the ratio of the total volume of the \(i\)th group bubbles per unit volume to the local volume fraction.

\[
\alpha_s f_i = n_i v_i \quad (4.23)
\]

where \(n_i\) is the number density of the \(i\)th group bubbles and \(v_i\) is their mean volume. The interfacial force \(\vec{F}_{lg}\) generally includes the forces due to viscous drag \(\vec{F}_D\), the lateral lift \(\vec{F}_L\), the wall lubrication \(\vec{F}_W\), and the turbulent dispersion \(\vec{F}_{TD}\), which are defined by the following equations:

\[
\vec{F}_{lg} = -\vec{F}_{gl} = \vec{F}_D + \vec{F}_L + \vec{F}_W + \vec{F}_{TD} \quad (4.24)
\]

\[
\vec{F}_D = \frac{3}{4} \frac{C_D}{d_b} \alpha_s \rho_i \left| \vec{U}_g - \vec{U}_l \right| \left( \vec{U}_g - \vec{U}_l \right) \quad (4.25)
\]

\[
\vec{F}_L = C_L \alpha_s \rho_i \left( \vec{U}_g - \vec{U}_l \right) \times \left( \nabla \times \vec{U}_i \right) \quad (4.26)
\]

\[
\vec{F}_W = -\frac{\alpha_s \rho_i \left( U_g - U_l \right)}{d_b} \max \left( 0, C_{w1} + C_{w2} \frac{d_b}{y_W} \right) \vec{n} \quad (4.27)
\]
\[ F_{TD} = -C_{TD} \rho k_i \nabla \alpha_i \] (4.28)

where \( d_b \) is the Sauter mean diameter of bubbles defined by:

\[ d_b = \frac{1}{N} \sum_{i=1}^{N} \frac{f_i d_i}{d_i} \] (4.29)

The drag coefficient \( C_D \) in Equation 4.25 is usually calculated according to Ishii and Zuber (1979). The lift coefficient \( C_L \) in Equation 4.25 is estimated using the Tomiyama model (Tomiyama, 1998). The wall lubrication coefficients and the turbulent dispersion coefficient take value of \( C_{W1} = -0.01 \), \( C_{W2} = 0.05 \) and \( C_{TD} = 0.1 \), respectively.

\[ C_D = \begin{cases} 
\frac{24}{Re_b} & 0 < Re_b \leq 0.2 \\
\frac{24}{Re_b} (1 + 0.1 Re_b^{0.75}) & 0.2 < Re_b \leq 1000 \\
2 Eo^{0.3} & 1000 < Re_b 
\end{cases} \] (4.30)

\[ Re_b = \frac{\rho \left| \bar{U}_b - \bar{U}_l \right| d_b}{\mu_i} \] (4.31)

\[ C_L = \begin{cases} 
\min \left[ 0.288, \tanh(0.121 Re_b, f(Eo^*)) \right] & Eo^* \leq 4 \\
f(Eo^*) = 0.00105 Eo^3 - 0.0159 Eo^2 - 0.0204 Eo^* + 0.474 & 4 < Eo^* \leq 10 \\
-0.27 & 10 < Eo^* 
\end{cases} \] (4.32)

\[ Eo^* = \frac{g (\rho_i - \rho_g) d_H^2}{\sigma} \] (4.33)

where \( Re_b \) and \( Eo^* \) represent bubble Reynolds number and modified Eötvös number, respectively. In Equation 4.33, \( d_H \) is the maximum bubble horizontal dimension which is related to the bubble aspect ratio, \( E \) (Wellek et al., 1966).

\[ d_H = d_b \left( \frac{1}{E} \right)^{1/3} \] (4.34)

The empirical correlation of Wellek et al. (1966) is used to evaluate \( E \):

\[ E = \frac{1}{1 + 0.163 Eo^{0.757}} \] (4.35)

### 4.2.2.2 Population Balance Method
The bubble size distribution is modelled using the population balance equation:
\[
\frac{\partial n_i}{\partial t} + \nabla \cdot \left( \vec{U}_i n_i \right) = \left( \sum_{j=1}^{i} S_{ij} \right)
\] (4.36)

where \((\sum_{j=1}^{i} S_{ij})\) is a source term describing the bubble number density variations due to coalescence and break-up. The mass variation of the \(i^{th}\) group bubbles in Equation 4.20 can be calculated by:
\[
\left( \sum_{j=1}^{i} \Gamma_{ij} \right)_{i} = \left( \sum_{j=1}^{i} S_{ij} \right) \left( \frac{\alpha_s f_i}{n_i} \rho_s \right)
\] (4.37)
\[
\left( \sum_{j=1}^{i} S_{ij} \right) = B_C + B_B - D_C - D_B
\] (4.38)

where \(B_B\) and \(D_B\) are, respectively, the birth and death rates of the number density of the \(i^{th}\) group bubbles due to break-up, \(B_C\) and \(D_C\) are the birth and death rates due to coalescence. Experimental observation by Li et al. (2010) demonstrated that in the case of upward bubbly flows in small-diameter vertical tubes, coalescence is predominant while break-up is almost invisible. Therefore, only the coalescence mechanisms are included in this study while bubble break-up is neglected.

\[
B_C = \frac{1}{2} \sum_{k=1}^{i} \sum_{j=1}^{i} n_i n_j \chi_{ij}
\] (4.39)
\[
D_C = \sum_{j=1}^{N} n_i n_j \chi_{ij}
\] (4.40)

\(\chi_{ij}\) in above equations is the coalescence rate of two bubble groups. According to the film drainage theory proposed by Shinnar and Church (1960), the coalescence of two bubbles occurs in three steps: (1) two bubbles collide, trapping a liquid film between them; (2) bubbles keep in contact while the liquid film drains; (3) when the contact time is sufficient for the liquid film to drain out down to a critical thickness, the film ruptures, resulting in coalescence. It is worth noting that not all collisions lead to coalescence. The concept of collision efficiency \(\lambda_{ij}\) is thus introduced to account for the probability of bubble coalescence:
\[
\chi_{ij} = \theta_{ij} \lambda_{ij}
\] (4.41)
where $\theta_{ij}$ is the collision frequency. In a turbulent flow, the collisions between bubbles may be caused by a number of mechanisms such as turbulent fluctuation, laminar shear, wake entrainment, and buoyancy. In this study, the former three mechanisms are taken into account. The collision frequency $\theta_{ij}$ is therefore written as:

$$\theta_{ij} = \theta_{ij}^T + \theta_{ij}^{LS} + \theta_{ij}^{WE}$$

(4.42)

where $\theta_{ij}^T$, $\theta_{ij}^{LS}$ and $\theta_{ij}^{WE}$ represent the collision frequency due to turbulence, laminar shear and wake entrainment, respectively. $\theta_{ij}^T$ is defined by (cited in (Li et al., 2010)):

$$\theta_{ij}^T = \frac{\pi}{4} (d_i + d_j)^2 (u_{Ti}^2 + u_{Tj}^2)^{1/2}$$

(4.43)

$$u_{Ti} = \sqrt{2} \varepsilon_1 d_i^{1/3} u_{Ti}, u_{Tj} = \sqrt{2} \varepsilon_1 d_j^{1/3}$$

(4.44)

The frequency of shear-induced collisions $\theta_{ij}^{LS}$ is given by (cited in (Li et al., 2010)):

$$\theta_{ij}^{LS} = \frac{32}{3} (d_i + d_j)^3 \left| \frac{dU_i}{dR} \right|$$

(4.45)

When bubbles enter the wake region of a leading bubble, they will accelerate and may collide with the preceding one, resulting in bubble coalescence. This mechanism is accounted using the model proposed by Wang et al. (cited in (Li et al., 2010)):

$$\theta_{ij}^{WE} = K \Theta d_i^2 u_{si}$$

(4.46)

where $K$ is a constant (K=15.4), $u_{si}$ is the slip velocity defined by:

$$u_{si} = 0.71 \sqrt{gd_i}$$

(4.47)

The parameter $\Theta$ is introduced in consideration that only bubbles larger than $d_{cr}/2$ have a wake region effect for bubble coalescence.

$$\Theta = \begin{cases} 
(d_i - d_{cr}/2)^6 & d_i \geq d_{cr}/2 \\
(d_i - d_{cr}/2)^6 + (d_{cr}/2)^6 & d_i \geq d_{cr}/2 
\end{cases}$$

(4.48)

$$d_{cr} = 4 \left( \frac{\sigma}{g(\rho_l - \rho_s)} \right)^{1/6}$$

(4.49)

According to Coulaloglou (cited in (Li et al., 2010)), the collision efficiency $\lambda_{ij}$ is determined by the actual contact time $\tau_{ij}$ and the drainage time $t_{ij}$, which is the time required for the liquid film to thin down to a critical thickness.
\[ \lambda_{ij} = \exp\left(-\frac{t_{ij}}{\tau_{ij}}\right) \]  

(4.50)

To estimate the bubble contact time \( \tau_{ij} \) in a turbulent system, the correlation developed by Levich et al. (cited in (Li et al., 2010)) is widely used:

\[ \tau_{ij} = \frac{r_{ij}^{2/3}}{\varepsilon^{1/3}} \]  

(4.51)

\[ r_{ij} = \left(\frac{1}{2} \left( \frac{1}{r_i} + \frac{1}{r_j} \right) \right)^{-1} \]  

(4.52)

The drainage time \( t_{ij} \) is calculated according to Prince and Blanch (1990):

\[ t_{ij} = \left(\frac{r_{ij}^3 \rho_l}{16 \sigma}\right)^{1/2} \ln \frac{h_0}{h_f} \]  

(4.53)

### 4.2.3 Numerical Procedure

Park and Chang (2011) conducted isothermal bubbly flow experiments under the atmospheric pressure and room temperature (25 °C), using pure water and dilute Al\(_2\)O\(_3\)/water nanofluid (0.1 vol%), respectively. The test section was a vertically oriented acrylic pipe with an inner diameter of 15 mm and a length of 2.5 m. Liquid and air bubbles were mixed at the bottom of the test section using a bubble formation bed and driven by a pump to flow upward. A conductivity double-sensor void meter was mounted at a height of 1.75 m downstream of the bubble formation bed. Local two-phase flow parameters including the void fraction, the bubble diameter and the bubble velocity were measured. These parameters were utilized in this study for model validation.

Due to the axial-symmetry of the flow field, a sector-shaped column computational domain which was a quarter of the pipe was built, as illustrated in Figure 4.10. The domain was then discretised using structured meshes. Mesh sensitivity test provided that mesh independence was achieved at 63360 cells since a further increase of mesh density to 144000 cells just caused a small change (less than 1%) in the predicted air velocity. The inlet boundary condition was carefully set up according to the experimental conditions: the superficial velocities of liquid and air were set to be 2.8294 m/s and 0.1886 m/s, respectively; the initial volume fraction of air at the inlet
was estimated to be 0.062. In order that the process of bubble coalescence can be efficiently represented, bubbles ranging from 1.5 to 15 mm diameter were equally divided into 9 groups. The size range and centre bubble diameters of each group are shown in Table 1. Since the bubbles in the experiments were injected through small holes (1 mm in diameter, 2 mm space from each other) on the bubble bed, the initial bubble size was estimated to be in the range of 1.5-3.0 mm, which completely fell in the 1st size group in the computations.

Figure 4. 10: The computational domain.

The water properties were referred to the water data from USGS, while the nanofluid properties such as density, viscosity and surface tension were calculated using the widely validated correlations in the literature (Wang and Bao, 2009; Khanafer and Vafai, 2011). Since the density of air is much lower than that of water and nanofluid, it was assumed that the motion of air bubbles follows the fluctuations in the continuous liquid phase. Thus the gas phase was assumed as laminar and the turbulence was modelled only for the liquid phase using the improved \( k-\varepsilon \) model of Sato and Sekoguchi (1975) in order to take into account the extra turbulence in the liquid phase induced by the bubbles. The conservation equations were then solved using the
commercial CFD code CFX-14.5. Convergence was achieved within 4000 iterations when the mass residual of the continuous phase dropped down to $1 \times 10^{-4}$.

### 4.2.4 Results and Discussion

#### 4.2.4.1 Comparison of simulation results against experimental data

The measured and predicted radial profiles, at $H = 1.75$ m, of void fraction, gas velocity, IAC and Sauter mean bubble diameter distribution in the air-water bubbly flow are depicted in Figure 4.11. Different mechanisms of bubble collision are represented by “T” “LS” and “WE”, for turbulent fluctuation, laminar shear and weak entrainment, respectively. As shown in Figure 4.11, when only the turbulent fluctuation induced collision was considered, the predicted void fraction and IAC profiles exhibited a wall-peaked shape which was totally different from the centrally distributed experimental data. Meanwhile, the Sauter mean bubble diameter was considerably under-predicted. When the laminar shear induced collision was added, although the model provided closer predictions, but the void fraction and Sauter mean bubble diameter were still underestimated. When all of the three mechanisms were taken into account, all the model predictions were in satisfactory agreement with the experimental measurements, which clearly demonstrated the importance of complete inclusion of collision mechanisms in the MUSIG model.

The predicted local flow parameters in the air-nanofluid bubbly flow were also compared against the experimental data, as shown in Figure 4.12. The experimental observation proved the addition of nanoparticles into the liquid had the tendency to flatten the radial distributions of void fraction, gas velocity and Sauter mean bubble diameter, but to increase the IAC. Unfortunately, the MUSIG model failed to capture these features. The void fraction, gas velocity and Sauter mean bubble diameter were all grossly over-predicted, while the IAC was considerably underestimated. The classic MUSIG model which has been successfully employed in air-water bubbly flows was proven inapplicable to nanofluids.
Figure 4. 11: Comparison of predicted flow parameters against experimental data of the air-water bubbly flow: (a) void fraction; (b) gas velocity; (c) IAC; (d) Sauter mean bubble diameter (Park and Chang, 2011).

One plausible explanation for the discrepancies in nanofluid case could be due to the over-prediction of the bubble size (shown in Figure 4.12(d)). Cheung et al. (2007) found that the over-predicted bubble size introduced significant error in the predicted void fraction, IAC and gas velocity. In fact, in bubbly flows the Sauter mean bubble diameter is generally closely coupled with the interfacial forces. This coupled system strongly affects the phase distribution patterns by influencing the transverse motion of bubbles in the liquid. As aforementioned, most of the closure equations related to bubble diameters and interfacial forces were empirically correlated to the experimental
data of pure liquids, it is therefore necessary to examine their applicability in air-nanofluid bubbly flows.

Figure 4. 12: Comparison of predicted flow parameters against experimental data of the air-nanofluid bubbly flow: (a) void fraction; (b) gas velocity; (c) IAC; (d) Sauter mean bubble diameter (Park and Chang, 2011).

4.2.4.2 Model Improvement for the effects of nanoparticle self-assembly

Nanoparticle self-assembly

The inherent instability of bubbles arises from the high free energy of the gas-liquid interface. Under the driving force of minimized interfacial energy, nanoparticles tend to spontaneously assemble at the gas-liquid interface (Sun et al., 2015) and forme a close-
packed particle layer. Shown in Figure 4.13 is a microscope image of the air bubbles surrounded by a thin layer of nanoparticles in the nanofluid (MAGSILICA® H8 nanoparticles at $C_p=10$ mg/mL in ethanol/water mixtures), as observed by Rodrigues et al. (2011). This phenomenon has been widely found in numerous experimental studies. Blute et al. (2009) even utilize this spontaneous nanoparticle assembly to prepare Langmuir-Blodgett films (one or more monolayers of nanoparticles) at the air-water interface. It is therefore reasonable to extrapolate this close-packed nanoparticle layer at the gas-liquid interface generates a sort of ‘colloidal armour’ which might change the characteristics of bubble surface.

Figure 4. 13: Transmission Electron Microscopy (TEM) image of air bubbles surrounded by MAGSILICA® H8 nanoparticles ($C_p=20$ mg/mL) in ethanol/water mixture (Rodrigues et al., 2011).

The drag force

For bubbles submerged in continuous liquid, Clift et al. (cited in (Dijkhuizen et al., 2010a)) found that the assembly of contaminants would change the slip condition at the interface from free-slip to no-slip, resulting in the increase of drag force (shown in Figure 4.14).

This is consistent with the hypothesis proposed by McClure et al. (2014) who experimentally proved when impurities were added to an air-water bubbly flow, the overall holdup would be changed. Similarly, they attributed this unique phenomenon to the increased drag force induced by the assembly of impurities at the air-water interface. In order to capture the hydrodynamic behaviours of bubbly flows with surfactants,
McClure et al.’s (2015) improved the classic Grace drag model by introducing an empirical constant ($k_s = 1.6$~$2.2$) to account for the effects of accumulated surfactants on drag enhancement:

$$C_{D,grace}' = k_s C_{D,grace} f(\alpha)$$  \hspace{1cm} (4.54)

where $C_{D,grace}$ is the drag coefficient calculated by the Grace drag model:

$$C_{D,grace} = \frac{4 g d_b}{3 U_T^2} \frac{(\rho_f - \rho_g)}{\rho_g}$$  \hspace{1cm} (4.55)

Figure 4.14: The effect of contaminants: (a) ultra-pure liquid with free-slip boundary condition; (b) slightly contaminated liquid with a limited circulation inside the bubble; (c) fully contaminated bubble with no-slip boundary condition (Dijkhuizen et al., 2010a).

As nanoparticles have comparable sizes with the micelles which are the aggregate of surfactant molecules ranging from 2 nm to 20 nm (Hasko, 1980), and also assemble at the interface in an analogous way, it is reasonable to hypothesize that the assembly of nanoparticles may have similar impact on drag enhancement. Following the modified Grace model by McClure et al. (2015), this study introduced the empirical constant ($k_s = 1.6$~$2.2$) to the Ishii-Zuber model (Ishii and Zuber, 1979) (Equation 4.30) to account for the influence of nanoparticles.

$$C_{D,ishii}' = k_s C_{D,ishii}$$  \hspace{1cm} (4.56)

The drag coefficients calculated before and after the modification are shown in Figure 4.15. The predicted void fraction and gas velocity with different $k_s$ values are compared in Figure 4.16. It clearly shows flattened distributions for both void fraction and gas velocity with the increasing drag force, which is consistent with experimental
measurements in contaminated air-water systems (Clift et al., 1978; McClure et al., 2015). The prediction of void fraction agreed reasonably well with the experimental data when $k_s$ took the value of 2.2, which evidently confirmed the effects of nanoparticle assembly on drag enhancement.

![Graph showing drag coefficient comparison](image)

**Figure 4. 15:** Comparison of predicted drag coefficients ($\sigma=0.065$ N/m, $\alpha_g=0.1$).

![Graph showing gas velocity and void fraction](image)

**Figure 4. 16:** Comparison of predicted flow parameters against experimental data of the air-nanofluid bubbly flow: (a) void fraction; (b) gas velocity (Park and Chang, 2011).
The lift force

The lift force, which acts in the direction perpendicular to the bubble movement and causes transverse bubble motion, is also altered by the assembled nanoparticles. According to Tomiyama (1998), the lift force would change its acting direction with increasing bubble sizes. Bubbles smaller than the critical diameter, which is 5.8 mm according to Tomiyama (1998), will be pushed by the positive lift force towards the wall, while those larger than 5.8 mm will move towards the tube axis under the action of negative lift force. Figure 4.12(d) shows that most bubbles in Park and Chang’s (2011) air-nanofluid bubbly flows were sized between 2 mm and 5 mm. According to Equation 4.32, a positive lift force pointing towards the wall would be generated, leading to a near-wall peaked void fraction distribution. Whereas, the measured void fraction has a core-peaked shape (Figure 4.12(a)), demonstrating the inapplicability of Equation 4.32 to bubbles in nanofluids.

For the air-nanofluid bubbly flows with $d_b=3$ mm (mean bubble diameter estimated according to Park and Chang (2011)), a satisfactory agreement was achieved when the lift coefficient took the value of $C_L = -0.025$ (Figure 4.17).

![](image)

Figure 4. 17: Predicted void fraction of the air-nanofluid bubbly flow with $C_L=-0.025$.

This indicates that the positive-to-negative transition in the lift coefficient occurs at a smaller bubble size in nanofluid than in water. According to Equation 4.33 and 4.34, the transition point is strongly affected by the bubble aspect ratio $E$. However,
due to the extreme complexity, $E$ is generally estimated empirically, such as the Wellek correlation (Wellek et al., 1966) (Equation 4.35) and the Okawa correlation (Okawa et al., 2003) (Equation 4.57):

$$E = \frac{1}{1 + 1.97Eo^{1.3}}$$ \hspace{1cm} (4.57)

For bubbles in nanofluids, the situation is even more complex because $E$ could be subject to many unknown mechanisms. Therefore, in this study, $E$ was re-defined according to Equation 4.58 in order to make the positive-to-negative transition of the Tomiyama lift coefficient occur at $d_b=2.9$ mm, as shown in Figure 4.18.

$$E = \frac{1}{1 + 6.099Eo^{1.3}}$$ \hspace{1cm} (4.58)

![Graph showing lift coefficient vs bubble diameter with three different correlation equations: Eq. 4.35, Wellek 1966; Eq. 4.57, Okawa 2003; Eq. 4.58, Present study. The graph displays a reversal point indicating a change in lift coefficient sign.]

Figure 4. 18: Comparison of predicted lift coefficients with different correlations of bubble aspect ratio.

**Reduced bubble coalescence rate induced by prolonged bubble drainage time**

It is interesting to note that the addition of nanoparticles leads to a decreased mean bubble size. As shown in Figure 4.12(d), most of the measured bubble diameters in nanofluids were between 2mm to 5mm, which were much smaller than those ranging from 3 mm to 10 mm in water (shown in Figure 4.11(d)). Analogous phenomenon was also reported by McClure et al. (2014) who observed a reduction in the mean bubble size in solutions with surfactants. They suggested those surfactants at air-water interface are effective in inhibiting bubble coalescence and thus responsible for the
smaller bubble size. Actually, nanoparticles were found to act in many ways like surfactant molecules, particularly if adsorbed to the interface (Binks, 2002). It is thus reasonable to extrapolate the layer of assembled nanoparticles at the bubble surface might play a similar role in reducing coalescence rate and impeding smaller bubbles coalescing into larger bubbles (Kam and Rossen, 1999). The reduced coalescence rate is perhaps the reason responsible for the smaller air bubble size in the Al₂O₃/water nanofluid as observed by Park and Chang (2011). As aforementioned, the coalescence rate \( \chi_{ij} \) depends on the collision frequency \( \theta_{ij} \) and collision efficiency \( \lambda_{ij} \). According to Equation 4.50, the collision efficiency is determined by the relative magnitude of contact time \( \tau_{ij} \) and drainage time \( t_{ij} \). Chesters (1991) pointed out that in fluid-liquid dispersions collision force and duration of two dispersed particles is only controlled by the external flow in the bulk. Thus the nanoparticles at the interface would probably not influence the collision frequency or contact time of two colliding bubbles, but elongate the drainage time. This hypothesis is also in line with the study by Kam and Rossen (1999) who found adsorbed solid particles at the gas-liquid interface can slow down film thinning by hindering the water flow at bubble surface. In order to take this effect into account, a correction coefficient \( k_d \) ranging from 1.0 to 2.0 was introduced to the Prince and Blanch model (Equation 4.53).

\[
\dot{t}_{ij} = k_d \cdot t_{ij} \text{(Prince–Blanch)}
\]  

(4.59)

Figure 4.19: Predicted film drainage time of equal size bubbles (\( \varepsilon=0.65 \text{ m}^2/\text{s}^3 \)).
The predicted bubble drainage time and collision efficiency using Equation 4.59 are illustrated in Figure 4.19 and Figure 4.20, respectively.

![Graph showing coalescence efficiency vs bubble diameter for different values of $k_d$.]

Figure 4.20: Predicted collision efficiency of equal size bubbles ($\varepsilon=0.65$ m$^2$/s$^3$).

With the increasing of $k_d$, the bubble drainage time increases gradually, while the collision efficiency decreases dramatically. Incorporating Equation 4.59 into the present MUSIG model with $k_d = 1.02$, the model achieved close predictions of void fraction with the experiment data, as shown in Figure 4.21. Moreover, from the phenomenological point of view, the phase distribution patterns along the radial direction of the pipe gradually changed from “core peak” ($k_d = 1.0$ and 1.02), to “transition” ($k_d = 1.06$), then to “intermediate peak” ($k_d = 1.1$), and finally to “wall peak” ($k_d = 2.0$) (Serizawa and Kataoka, 1988). Figure 4.22 depicts the predicted bubble size distribution at $H = 1.75$ m. Most bubbles fell in group 4 (4.5~6 mm), when $k_d$ ranged from 1.0 to 1.06. However, the largest proportion of bubbles moved to group 2 (1.5~3 mm) for $k_d = 2.0$ corresponding to the wall peak in Figure 4.21.

The above numerical results demonstrate that the appropriately formulated bubble drainage time is crucial to the prediction of Sauter mean bubble diameter and void fraction. However $k_d$ is case sensitive and subject to a number of factors including the nanoparticle material, size and concentration, as well as the flow conditions. In order to achieve a mechanistic modelling of bubble coalescence in nanofluids, further in-depth investigation is in urgent demand.
4.2.4.3 Effects of Nanoparticle Self-assembly on Liquid Film Drainage

It is widely believed that the driving force for nanoparticle assembly is the spontaneously reduced interfacial energy. The transmission electron microscopy (TEM) images obtained by Böker et al. (2007) demonstrates that the assembly process includes three steps: firstly, free nanoparticles diffuse to the interface; secondly, the particles
pack closer and form clusters which grow to form a closely packed particle array, lowering the interfacial tension, and; finally, thermally activated exchange between adsorbed and incoming particles is observed, leading to a tightly packed monolayer. This layer of adsorbed particles at the interface was found to be able to stabilize gas bubbles in liquid and has been widely employed to fabricate liquid foams with fine textures (Worthen et al., 2013).

One of the possible reasons leading to the stabilization could be the altered bubble surface properties. According to Lee and Hodgson (1968), the film drainage which dominates the bubble coalescence process is strongly affected by the rigidity of bubble surfaces (deformable and non-deformable) and the mobility of the contact interfaces (immobile, partially mobile and fully mobile). Tomiyama et al. (1998) proposed that the accumulation of impurities on a bubble surface will cause the interface to behave like a rigid surface. Worthen et al. (2013) suggested the addition of nanoparticles could increase the effective viscosity of the injected gas in the liquid and thereby reduce the bubble mobility. When the mobility of the bubble surface is restricted, the thinning process will be controlled by viscous effects and occur much slower, which will effectively prevent small bubbles coalescing into larger bubbles. This was perhaps the reason why Park and Chang (2011) observed smaller bubbles in the Al₂O₃/water nanofluid.

According to Oolman and Blanch (1986), the thinning of the liquid film trapped between two colliding bubbles is driven by the pressure forces. In the Prince and Blanch (1990) model (Equation 4.53), only the capillary pressure, which is induced by variations in the curvature of the gas-liquid interface, is included to account for the bubble drainage time.

\[ \Pi_c = \frac{2\sigma}{r^*_g} \]  

(4.60)

For pure liquids, it is true that the capillary pressure is the only force acting on the liquid film. But when a second component exists in the liquid, other forces resisting the film thinning can develop and elongate the bubble drainage time (Oolman and Blanch, 1986). Thus these disjoining forces induced by nanoparticles at the interface were perhaps another reason for the smaller and more stable bubbles in nanofluids.
Oolman and Blanch (1986) proposed that the surface activity of the second component can induce a surface tension force resisting the approach of two colliding bubbles. When the liquid film thins, the surface area increases. As a result, the surface concentration of adsorbed surfactants decreases. Since surface tension is an inverse function of surfactant’s concentration, a surface tension gradient along the thinning film will develop (Figure 4.23), resulting in an additional surface tension force. As proven by Böker et al. (2007), nanoparticles and surfactants have similar effects on lowering surface tension. When nanoparticles exist at the interface, the surface tension gradient and the surface tension force might be introduced to the bubbles as well. The net surface tension force along the radial dimension of the film could be expressed as follows (cited in (Oolman and Blanch, 1986)):

$$\Delta \sigma = \frac{1}{h} \left( \frac{2c}{RT} \right) \left( \frac{\partial \sigma}{\partial c} \right)^2$$

(4.61)

where $h$, $c$, $R'$, and $T$ represent film thickness, solute concentration, ideal gas constant, and absolute temperature, respectively.

Figure 4.23: The surface tension gradient along the radial dimension of the liquid film.

In addition, Wang and Yoon (2008) believed that the electrostatic double layer force plays a significant role in preventing the thinning of liquid film. The surface of air bubbles was found to acquire a negative charge in distilled water (Elmahdy et al., 2008) over the most of the pH range. The electrostatic double layer would thus be established in the bubbly flows. Due to their dielectric properties, non-metallic nanoparticles in electrostatic double layers will be polarized and charged (Marek et al., 2010). In turn, the charged nanoparticles will affect the surface charge density of the bubbles (Wu et al., 2015), changing repulsive electrostatic double layer force between two negative-charged bubbles (Figure 4.24). Consequently, the film thinning process would be
slowed down. The equation of electrostatic double layer force is usually given by (Bhattacharjee et al., 1998):

\[ \Pi_e = \left( 64 \pi k_B T r_h \rho_\infty \gamma^2 / \kappa^2 \right) \exp(-\kappa h) \]  

(4.62)

where \( k_B \), \( r_h \), \( \rho_\infty \), \( \gamma \), \( \kappa \) and \( h \) represent the Boltzmann constant, bubble radius, density of electric charge in the bulk solution, reduced surface potential, Debye screening length and film thickness, respectively.

Figure 4. 24: The electrostatic double layer force between two negative-charged bubbles.

Besides the aforementioned hypotheses, Samanta and Ghosh (2011) proposed that the reduced bubble coalescence in contaminated systems is mainly due to the steric force imparted by the adsorption of amphiphilic contaminants at air-water interface. The adsorbed layer encounters a reduction in entropy when confined in a very small space as the bubble approaches to each other. Since the reduction in entropy is thermodynamically unfavourable, their approach is thus inhibited. According to Böker et al. (2007), some nanoparticles such as Janus-particles like polymers have two surface regions: polar surface region and apolar surface region. These nanoparticles are surface active and amphiphilic (Böker et al., 2007). It is reasonable to extrapolate that when two bubbles approach to each other, similarly to the polymeric surfactant, the hydrated head groups of adsorbed nanoparticles will be overlapped, generating a steric repulsion force. This force could be calculated by (cited in (Samanta and Ghosh, 2011)):

\[ \Pi_s = \frac{k_B T}{s^3} \left[ \left( \frac{2L}{\delta} \right)^{9/4} - \left( \frac{\delta}{2L} \right)^{3/4} \right] \]  

(4.63)

where \( \delta \), \( L \), \( s \) represent the separation between the surfaces, the thickness of the polymer layer, the mean distance between the attachment points.

All of these three hypotheses are based on the nanoparticle layer at the interface. Therefore, in order to propose a mechanistic model for the bubble drainage time, more
details of the structure of nanoparticles at the interface are needed. In recent years, a few factors including the nanoparticle aspect ratio, surface properties, concentration and solvent evaporation rates were revealed to affect the orientation and packing structures of nanoparticles at the interface (Böker et al., 2007). Further studies showed that temperature, pH, base liquid polarity and redox activity could all control the interactions between particles and influence the structure of assembled nanoparticles (Marek et al., 2010). Moreover, for nanoparticles ranging from 10 nm to 1 μm, their assembly even could be directed by macroscopic viscous flows (Marek et al., 2010). In view of these novel experimental findings, it is obvious that the more details are uncovered, the more complex the problem will be. Due to the inherent complexity, substantial fundamental studies still need to be conducted in the future.

4.2.5 Conclusions

The MUSIG model was employed in this study to simulate air-water and air-nanofluid bubbly flows in a vertical tube under isothermal conditions. It was found that the classic MUSIG model achieved satisfactory agreement with the experimental data of air-water bubbly flow, whereas notable discrepancies were observed in the case of air-nanofluid bubbly flow. Based on the analysis of the numerical results, some potential mechanisms possibly responsible for the significantly changed two-phase flow structures were discussed and recommendations for future work were given. The conclusions arising from this study are as follows:

(1) The spontaneous assembly of nanoparticles at the bubble interface significantly changes the interface rigidity and mobility. As a result, the interfacial drag force is increased and the role of lift force with increasing bubble size is modified. It was proven that the positive-to-negative reversal of the lift force occurs at a smaller bubble size in nanofluids compared to that in pure water.

(2) The layer of nanoparticles at the bubble surface hinders bubble coalescence by forming a physical barrier and restricting the mobility of the surface. The thinning process of the liquid film trapped between two colliding bubbles slows down, resulting in a longer bubble drainage time. However, the mechanisms responsible for the elongated drainage time are still yet to be uncovered.
Chapter 5

Mechanistic Study of Bubble Hydrodynamics in Nanofluids

The main findings of this chapter have been included in:


5.1 Mechanistic Analysis of the Effects of Nanoparticles on Interfacial Forces on Bubbles in Nanofluids

Abstract

Bubbly flows of air-water and air-nanofluid were investigated numerically using the two-fluid model. Through comparing the predicted bubble velocity and void fraction profiles against the experimental data, the classic two-fluid model, which has been widely validated for two-phase flows of pure liquids, was found to be inapplicable to those of nanofluids because of the empirical nature of the interfacial force formulation. The roles of interfacial forces were believed to be significantly altered in nanofluids rather than in pure liquids due to the spontaneous phenomenon of nanoparticle adsorption at bubble interfaces. Because of the nanoparticle layer, bubbles submerged in nanofluids would partially behave like a rigid sphere and develop a rotation movement. A slanted wake could be induced behind the bubble, generating a lateral Magnus force pointing towards the pipe centre and consequently making the positive-to-negative reversion of lift force occur at a smaller bubble diameter. Meanwhile, the slanted wake would also make bubbles in the viscous regime experience a drag force similar to that in the distorted regime, which makes the viscous-to-distorted transition point occur at a smaller bubble Reynolds number. It was recommended that the most important task when modelling bubbly flows of nanofluids using the two-fluid model is to reformulate the interfacial forces accounting for the effects of nanoparticle adsorption.

5.1.1 Introduction

Heat transfer enhancement has long been a hot research topic because of the continuously increasing demands for heat removal in many industries. Thanks to the development of nano-technology, a new type of engineered colloidal dispersions of nanometre-sized particles in common base liquids, the so-called “nanofluid”, have been regarded as a revolutionary heat transfer medium in view of its significant heat transfer enhancements in nucleate boiling (Yang and Liu, 2011; Sheikhbahai et al., 2012; Kamatchi and Venkatachalapathy, 2015). As the formation of a thin layer of deposited
nanoparticles on the heater surface was widely observed in most nucleate boiling experiments using nanofluids, which does not exist in nucleate boiling of pure liquids, the heat transfer enhancement of nanofluids has been generally attributed to the surface modification induced by nanoparticle deposition during the boiling process (Ahmed and Hamed, 2012). It is believed that the deposited nanoparticles play a dominant role in altering the boiling heat transfer intensity through significantly changing the microstructures and properties of the heater surface, as well as the characteristics of bubble dynamics (Vafaei and Borca-Tasciuc, 2013). In the meantime, nanofluids for heat transfer applications are generally dilute with every low nanoparticles loads (less than 0.1 vol%). Under such low nanoparticle concentrations, the liquid thermophysic properties are negligibly modified (Kim, 2009), which makes it safe to assume that dilute nanofluids behave hydrodynamically identical to their pure base liquids. Some numerical studies on boiling flows of nanofluids further assumed the two-phase behaviours of nanofluids are also identical to those of base liquids (Li et al., 2014b), and only focused on the effects of surface modifications induced by nanoparticle deposition.

However, emerging evidence in recent year revealed that nanoparticles have significant impact on the two-phase flow structures and dynamics. Using a high-speed visualization and image processing technology, Rana et al. (2014) measured the void fraction in boiling flows of water and ZnO/water nanofluids (0.001~0.01 vol%) in horizontal annulus. They found that with the increasing nanoparticle concentration, the void fraction in nanofluid decreased as much as 86% when compared to that in water, which indicates that ZnO nanoparticles in fluid act as void-fraction-suppressing agent. The hydrodynamic behaviours in the pool boiling of water and Al2O3/water nanofluids (0.001 and 0.002 vol%) were also investigated by Dominguez-Ontiveros et al. (Dominguez-Ontiveros et al., 2010). Through comparing the velocity profiles obtained by Dynamic Particle Image Velocimetry (DPIV), the fluid velocity distributions were found to be generally less uniform and lower in magnitude for the nanofluid cases than for those of the pure water case. Recently, radial distributions of air-nanofluid (0.1 vol% Al2O3/water) bubbly flow parameters in a vertical tube were measured by Park and Chang (2011). The measurements showed that the air-nanofluid bubbly flow had a
more flattened void fraction distribution, lower bubble velocity, higher interfacial area concentration and smaller bubble size than those in the air-water flow.

Considering the profound inter-coupling of two-phase flow structures and the overall heat transfer performance (Atmane and Murray, 2005), it is crucial to achieve an effective modelling of the two-phase flow dynamics in order to obtain comprehensive predictions of nanofluid boiling flows in the future. Beyond that, as nanoparticles are finding an increasing number of applications in various industries, multi-dispersed bubbly systems containing nanoparticles are commonly encountered. For example, nanoparticles are tested at the laboratory scale in bubble column reactors (Abkarian et al., 2007) to enhance chemical reactions and interfacial mass transfer, and they are also used as surfactants to stabilize emulsions (Dickinson, 2010) and foams (Sun et al., 2015). An in-depth understanding of the effects of nanoparticles on bubble behaviours in liquids is obviously beneficial to many emerging and traditional industries.

Therefore, this study tries to reveal the mechanistic effects of nanoparticles on two-phase flow dynamics, with the aim to improve the two-fluid model for effective modelling of bubbly flows of nanofluids with and without heat and mass transfer.

5.1.2 Theoretical Models

Numerous studies (Palm et al., 2006) have demonstrated that due to their small sizes, nanoparticles could be assumed to be mixed with the base fluid at a near-molecular level and thus a nanofluid can be numerically treated as a pseudo-homogeneous single-phase liquid. The framework of the two-fluid model (Ishii, 1975), which has been regarded as the mechanistic macroscopic formulation of the thermal-hydraulic dynamics of two-phase flow system, is theoretically applicable to bubbly flows of nanofluids. In the model, two sets of conservation equations governing the balance of mass, momentum and heat of gas and liquid are solved. For an isothermal air-nanofluid flow, the two-fluid model takes the following form:

The continuity equation:

\[
\frac{\partial}{\partial t} \left( \alpha_k \rho_k \right) + \nabla \cdot \left( \alpha_k \rho_k \vec{U}_k \right) = 0
\]  
(5.1)
The momentum equation:
\[
\frac{\partial}{\partial t}\left(\alpha_k \rho_k U_k\right) + \nabla \cdot \left(\alpha_k \left(\rho_k U_k U_k - \mu_k \left(\nabla U_k + \left(\nabla U_k\right)^T\right)\right)\right) = \alpha_k \left(B - \nabla P_k\right) + \bar{F}_k
\]
(5.2)

where the subscripts \(k\) is the phase denotation (\(k=l\) for the liquid phase and \(k=g\) for the gas phase); \(\alpha, \rho, B, \bar{U}\) and \(\bar{F}_k\) represent the volume fraction, density, body force, velocity and interfacial forces, respectively.

For bubbles submerged in a continuous liquid, the interfacial force \(\bar{F}_k\) generally includes the forces due to drag and the effects of lateral lift, wall lubrication and turbulent dispersion.

\[
\bar{F}_k = -\bar{F}_g = \bar{F}_D + \bar{F}_L + \bar{F}_{TD} + \bar{F}_W
\]
(5.3)

The drag force \(\bar{F}_D\) is calculated by:

\[
\bar{F}_D = \frac{3}{4} C_D \alpha \rho_l |\bar{U}_g - \bar{U}_i| \left(\bar{U}_g - \bar{U}_i\right)
\]
(5.4)

The drag coefficient \(C_D\) is empirically correlated by Ishii and Zuber (1979) to the bubble Reynolds number \(Re_b\) and Eötvös number \(Eo\):

\[
C_D = \begin{cases} 
24 / Re_b & 0 < Re_b \leq 0.2 \\
24(1 + 0.1 Re_b^{0.73}) / Re_b & 0.2 < Re_b \leq 1000 \\
2 Eo^{0.5} / 3 & 1000 < Re_b
\end{cases}
\]
(5.5)

\(Re_b\) and \(Eo\) are defined by:

\[
Re_b = \frac{\rho_g |\bar{U}_g - \bar{U}_i| d_b}{\mu_l}
\]
(5.6)

\[
Eo = \frac{g(\rho_i - \rho_g)d_b^2}{\sigma}
\]
(5.7)

When a bubble moves in a liquid, it experiences a transverse force which is usually called the lift force \(\bar{F}_L\). The general form of the lateral lift force is given by Drew and Lahey (cited in (Kolev, 2012)):

\[
\bar{F}_L = C_L \alpha \rho_l |\bar{U}_g - \bar{U}_i| \times \nabla (\bar{U}_g - \bar{U}_i)
\]
(5.8)
The empirical Tomiyama correlation (Tomiyama et al., 2002) is generally used to calculate the lift coefficient $C_L$:

$$
C_L = \begin{cases} 
\min \left[ 0.288, \tanh(0.121Re_s, f(Eo^*)) \right] & Eo^* \leq 4 \\
f(Eo^*) = 0.00105Eo^3 - 0.0159Eo^2 - 0.0204Eo^* 0.474 & 4 < Eo^* \leq 10 \\
-0.27 & 10 < Eo^* 
\end{cases}
$$

(5.9)

where $Eo^*$ is the modified Eötvös number based on the maximum bubble horizontal dimension $d_H$ (Wellek et al., 1966).

The wall lubrication force $\bar{F}_W$ and the turbulent dispersion force $\bar{F}_{TD}$ were calculated by Equation 5.10 and Equation 5.11, respectively (cited in (Kolev, 2012)).

$$
\bar{F}_W = -\frac{\alpha_s \rho_l (\bar{U}_g - \bar{U}_l)}{d_b} \max \left( 0, C_{w1} + C_{w2} \frac{d_n}{y_w} \right) \tilde{n} 
$$

(5.10)

$$
\bar{F}_{TD} = -C_{td} \rho_l k_l \nabla \alpha_i
$$

(5.11)

Due to the inherent complexity, the coefficients for interfacial forces are generally formulated empirically, or at least semi-empirically. Among them, the formulation of drag coefficient and lift coefficient has a significant effect on the overall modelling because bubble movement in liquid was reported to be largely controlled by the interaction between the drag force and the lift force. Although satisfactory predictions have been achieved using the Ishii-Zuber drag correlation (Ishii and Zuber, 1979) (Equation 5.5) and Tomiyama lift correlation (Tomiyama et al., 2002) (Equation 5.9) in a number of studies dealing with bubbly flows of pure liquids (Li et al., 2006), their applicability to nanofluids are still questionable due to the modifications induced by the existence of nanoparticles in the liquid.

In order to assess the validity of current “main-stream” interfacial force coefficients, Park and Chang’s (2011) experimental data of air-water and air-nanofluid (0.1 vol% Al$_2$O$_3$/water) bubbly flows in a vertical tube were employed in this study for model validation. The test section is a vertically oriented acrylic tube with an inner diameter of 15 mm and a length of 2.5 m. Nanofluid (0.1 vol% Al$_2$O$_3$/water) and air bubbles were mixed at the bottom of the test section through a bubble formation bed.
The mixture was then driven by a pump to flow upward. By controlling the superficial velocities at $j_l = 2.83 \text{ m/s}$ for the liquid and $j_g = 0.19 \text{ m/s}$ for the air, respectively, a stable bubbly flow was achieved. The two-phase flow parameters such as the void fraction, the bubble diameter and the bubble velocity were measured using a conductivity double-sensor two-phase void meter located 1.75 m downstream of the bubble formation bed.

The aforementioned model equations are solved using the commercial CFD code ANSYS CFX 16.0. In order to facilitate the comparison between numerical results and experimental data, the boundary conditions of the computations were carefully set up according to the experimental conditions. Details of the boundary conditions and numerical procedures have been highlighted in our previous studies (Li et al., 2016) and will not be repeated here. Uniform bubble diameters of $d_b = 6 \text{ mm}$ for the air-water case and $d_b = 3 \text{ mm}$ for the air-nanofluid case were estimated, respectively, based on the experimental data. The liquid properties of the Al$_2$O$_3$/water nanofluid were estimated using the correlations listed in Table 1. During the computations, the flow of the gaseous phase was assumed to be laminar as the air density is much lower than the liquid density and the motion of air bubbles follows the fluctuations in the continuous liquid phase. Turbulence was only modelled for the liquid phase using the improved $k$-$\varepsilon$ model by Sato and Sekoguchi (1975), which takes into account the effects of the bubble-induced additional turbulence viscosity on the liquid effective viscosity.

Table 5.1 Employed physical properties for mathematical modelling.

<table>
<thead>
<tr>
<th>Properties</th>
<th>Expression</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density</td>
<td>$\rho_{uf} = \rho_l (1 - \phi_v) + \rho_v \phi_v$</td>
<td>Based on the principle of the mixture rule; The validity has been examined by Park and Cho with water-Al$_2$O$_3$ nanofluid (0-5 vol%) (cited in Khanafer and Vafai, 2011)</td>
</tr>
<tr>
<td>Viscosity</td>
<td>$\mu_{uf} = (1 + 2.5\phi_v) \mu_l$</td>
<td>Proposed by Einstein based on the phenomenological hydrodynamic equations; valid for low concentration (0-2 vol%) (cited in Khanafer and Vafai, 2011)</td>
</tr>
<tr>
<td>Surface tension</td>
<td>$\sigma_{uf} = 0.065 N/m^2$</td>
<td>Based on the experiment of Wang and Bao (2009)</td>
</tr>
</tbody>
</table>
5.1.3 Results and Discussion

5.1.3.1 Comparison of the Numerical Results against Experimental Data

Figure 5.1: Comparison of predicted bubble velocity and void fraction profile against experimental data: (a) air-water bubbly flow; (b) air-nanofluid bubbly flow (Park and Chang, 2011).

The predicted bubble velocity and void fraction of the air-water and air-nanofluid bubbly flows were compared against the experimental data of Park and Chang (2011) in Figure 5.1, respectively. A well-developed core-peaking distribution of void fraction

145
was observed in the air-water bubbly flow, which was successfully captured by the two-fluid model (Figure 5.1(a)). In the case of air-nanofluid bubbly flow, the bubble velocity was suppressed and the overall shape of void fraction distribution was flattened (Figure 5.1(b)) when compared to the air-water case. The two-fluid model, however, overestimated the bubble velocity slightly and predicted a completely incorrect wall-peaking void fraction distribution for the air-nanofluid bubbly flow, indicating significant improvement is needed in order that a satisfactory prediction could be achieved.

5.1.3.2 The adsorption of nanoparticles on air-water interface

The experimental observation by Park and Chang (2011), among many others (Dominguez-Ontiveros et al., 2010; Rana et al., 2014), clearly demonstrated that the addition of nanoparticles into the base liquid induced a significant alteration in the two-phase flow structures and parameter profiles, even if the amount of added nanoparticle was so small (0.1 vol%) that the liquid properties were only negligibly changed. They attributed the modified flow dynamics of nanofluids to the altered interfacial drag and lift forces, but did not provide further mechanistic explanations for the questions like what role nanoparticles have played in altering the drag and lift forces, and how the altered drag and lift forces affect the structure of air-nanofluid bubbly flow. Experimental data from McClure et al.’s recent study (McClure et al., 2017) showed a similar effect of the surfactant addition on the hydrodynamics in a bubble column. Significant reductions in the average hold-up (up to 30%) and the Oxygen Transfer Rate (OTR, up to 75%) were observed with a small amount of surfactants (0.01 vol%). Such reductions are commonly attributed to the accumulation of the surfactants at the gas-liquid interface (McClure et al., 2017). Coincidently, as early as 2016 our previous study (Li et al., 2016) proposed a similar hypothesis that the adsorbed nanoparticles at the bubble interfaces seem to be a plausible cause responsible for the prominently modified two-phase flow features. However, the underlying mechanisms are still yet to be revealed.

In fact, the phenomenon of nanoparticle adsorption at phase interfaces has long been recognized and vastly investigated. Under the driving force of minimized Gibbs
free energy, nanoparticles suspending in the liquid tend to spontaneously aggregate at bubble interfaces (Sun et al., 2015). Shown in Figure 5.2 is an image of nanoparticle adsorption (MAGSILICA® H8 nanoparticles) at the interface of bubbles submerged in ethanol/water mixture (Rodrigues et al., 2011). Using the Confocal Laser Scanning Microscopy (CLSM) technology, Dickinson et al. (2004) observed a thin layer of silica nanoparticles surrounding air bubbles in the nanofluid (1 wt% silica nanoparticles in NaCl/water solutions) and further proved the existence of the close-packed particle layer around the bubble (Binks and Horozov, 2005).

![TEM image of air bubbles with MAGSILICA® H8 nanoparticles (Cₚ=10mg/mL) in ethanol/water mixtures (Rodrigues et al., 2011).](image)

Hunter et al. (2008) found that the detachment energy, which is related to the free energy required to remove an adsorbed nanoparticle from the interface, can be up to several thousand $k_BT$ ($k_B$ is the Boltzmann constant and $T$ is the absolute temperature), which is much higher than the detachment energy needed for surfactants. This means once nanoparticles are adsorbed, it is almost impossible to force them out of the bubble interface. Therefore, the layer of adsorbed nanoparticles at the gas-liquid interface generates a sort of “colloidal armour” that can inhibit, or even overwhelmingly stop smaller bubbles coalescing into larger bubbles (Kam and Rossen, 1999; Du et al., 2003). This was perhaps the reason why Park and Chang (2011) observed smaller bubbles in the $\text{Al}_2\text{O}_3$/water nanofluid than in pure water. It is also speculated that with this “colloidal armour”, the gas-liquid interface would behave pretty much like a rigid surface rather than a mobile one. Bubbles packed with a nanoparticle layer would be
consequently more like a rigid sphere than a deformable bubble. In addition, the slip condition of the bubble surface is also believed to be changed from free-slip to no-slip, as shown in Figure 5.3 (Dijkhuizen et al., 2010a), and consequently the inner circulation flow in bubbles is also partially or completely suppressed depending on the rigidity of the packed bubble. Therefore, the roles of interfacial forces, especially those of the lift force and drag force, would be significantly altered due to the modified bubble interface properties.

Figure 5.3: The effect of contaminants: (a) ultra-pure liquid with free-slip boundary condition; (b) slightly contaminated liquid with a limited circulation inside the bubble; (c) fully contaminated bubble with no-slip boundary condition (Dijkhuizen et al., 2010a).

5.1.3.3 Analysis of the Lift Force

The transverse motion of bubbles in a vertical flow is largely controlled by the lift force, which acts perpendicularly to the bubble rising direction. For small spherical bubbles in an upward flow, the lift force is mainly resulted from laminar shear and acts towards the pipe wall (Figure 5.4). The lift coefficient $C_L$ is positive with a value ranging from 0.25 to 0.5 (Zun, 1980; Auton, 1987; Lance and de Bertodano, 1994) depending on the liquid viscosity.

With increasing bubble size, bubbles tend to deform because of the free surface mobility and induce a wake behind the bubble, as shown in Figure 5.5. The wake is generally slanted due to the liquid velocity gradient. Through analysing the shape and trajectories of air bubbles rising in glycerol/water solutions with a video camera, Tomiyama et al. (1995) confirmed Serizawa and Kataoka’s (1994) presumption that the
lateral migration of a deformed bubble is governed by the complex interactions between the bubble wake and the liquid shear field. Based on their experimental observations, Tomiyama et al. (1995) proposed that the slanted wake can cause a lift force acting towards the pipe centre. When the wake becomes strong enough, the wake-induced lift force is able to defeat the shear-induced lift force and causes a lift force reversion. They also developed an empirical $C_L$ correlation (Equation 5.9) which has allowed modelling the transverse migration of spherical and deformed bubbles (Tomiyama et al., 2002).

Figure 5.4: Lift force on a spherical bubble in pure liquids.
However, when bubbles are covered with nanoparticles, the situation may be different. With the so-called “colloidal armour”, bubbles in nanofluids are partially rigid and more resistant to deform. Under the action of liquid velocity gradient, a nanoparticle-covered bubble tends to develop a rotating movement, which induces a slanted wake behind the bubble and generates a lateral force pointing towards the pipe centre, as illustrated in Figure 5.6. The lateral force induced by rotating spherical objects is well known as the Magnus force (Bagchi and Balachandar, 2002), which is essentially a wake-induced lateral lift force but expected to have a much stronger effect on bubble transverse migration than the deformation effect.

![Diagram](image)

Figure 5. 6: Lift forces on a nanoparticle-covered spherical bubble in nanofluids.
Figure 5. 7: Bubble lift coefficient versus bubble diameter.
Figure 5. 8: Predicted bubble velocity and void fraction profile of air-nanofluid bubbly flows with $C_L = -0.025$: (a) Void fraction; (b) Bubble velocity.

Therefore, the lift forces exerted on a deformed bubble in water and on a nanoparticle-covered bubble in nanofluids are both expected to be a consequence of two competing factors: shear- and wake-induced lift forces. In air-water bubbly flows, the shear effect is dominant for small spherical bubble with low Eötvös number and the lift coefficient is positive, as shown in Figure 5.7. As the bubble size increases, the wake effect due to the bubble deformation becomes increasingly important and finally reverses the sign of the lift coefficient at the critical bubble diameter, ($d_{cr} = 5.8\text{mm}$ according to Tomiyama correlation (Equation 5.9)). Therefore, the bubbles with an average diameter $d_b = 6\text{mm}$ in the air-water case of this study had a negative lift force and migrated towards the pipe centre, constituting the core peaking of the void fraction distribution shown in Figure 5.1(a).

In air-nanofluid bubbly flows, the sign of the lift force is less controlled by the Eötvös number because the bubbles are deformation-resistant, but is expected to be controlled by the bubble Reynolds number. According to Moraga et al. (1999), no wake-induced lift force is expected for Reynolds numbers below 300 where the lift coefficient is positive. As the Reynolds number increases, the wake effect due to the rotation becomes increasingly important and eventually reverses the sign of the lift coefficient to negative. According to Park and Chang’s experiment (2011), the bubble Reynolds number in the air-nanofluid bubbly flow was estimated to be 1000. Obviously,
the lift coefficient was negative even for the small spherical bubble. In fact, the negative lift force has been observed with surfactant-contaminated spherical bubbles by Fukuta et al. (2008) and with rigid spheres by Kurose and Komori (1999). In this study further simulations were conducted and it was found that when the lift coefficient took the value of $C_L = -0.025$, good agreement with the experiment data was achieved for the void fraction profile, despite the bubble velocity was still slightly over-predicted (Figure 5.8).

Since the average bubble diameter in air-nanofluid case was estimated to be 3mm in the air-nanofluid case in this study, it is reasonable to expect that the positive-to-negative transition occurred at a smaller critical bubble diameter than 3mm (Figure 5.7). However, the Tomiyama correlation (Equation 5.9) gives a critical bubble diameter of $d_{cr}=5.5\text{mm}$, which is much larger than the actual average bubble diameter, leading to a positive lift coefficient ($C_L=0.288$) and the incorrect prediction of the near-wall peaked void fraction profile (Figure 5.1(b)). Therefore, for bubbles in nanofluids the expected lift coefficient curve should locate left to the Tomiyama curve, as shown in Figure 5.7.

### 5.1.3.4 Analysis of the Drag Force

Since the drag force has strong effects on the rise velocity of bubbles, the slightly over-predicted bubble velocity (Figure 5.8(b)) is expected to be attributed to the altered drag force by the nanoparticle adsorption at bubble interfaces.

Clift et al. (1978) found the existence of surfactants at the bubble surface could increase the shear drag by changing the slip condition of the bubble surface from free-slip to no-slip and significantly hindering the internal circulation within the bubble (see Figure 5.3). Under a similar assumption, McClure et al. (2015) improved the classic Grace model by multiplying an empirical constant ($k_f=1.6\sim2.2$) to account for the effects of accumulated surfactants on the drag enhancement (McClure et al., 2014):

$$C_D^* = k_f C_{D,\text{Grac}} f(\alpha) \tag{5.12}$$

Similarly, Tomiyama et al. (1998) also proposed an empirical correlation to take account of the drag enhancement induced by the aggregation of contaminants at bubble interfaces.
\[ C_{T,Tomiyama} = \max \left[ 24(1 + 0.15Re_b^{0.687}) / Re_b, 8Eo / 3(Eo + 4) \right] \]  \hspace{1cm} (5.13)

As nanoparticles are found to behave like general surfactants in many ways like adsorbing at the bubble interfaces and changing the slip condition (Binks, 2002), it is extrapolated that adsorbed nanoparticles might play a similar role in increasing the shear drag. Following the modified Grace model, this study introduced the empirical constant \((k_s =1.6\sim2.2)\) to the Ishii-Zuber model (Ishii and Zuber, 1979) (Equation 5.5) to account for the influence of nanoparticles:

\[ C_D' = k_s C_{D,Ishii} \]  \hspace{1cm} (5.14)

![Figure 5.9: Comparison of predicted bubble velocity profiles using different drag correlations.](image)

For the purpose of comparison, the Tomiyama drag correlation (Equation 5.13) was also included in the computations. A comparison of numerical results against the experimental data is shown in Figure 5.9. It demonstrates that accounting for the influence of adsorbed nanoparticles on shear drag through a simple coefficient only has very limited impact on the bubble velocity prediction. This indicates that the effects of nanoparticles on bubble drag would be much stronger than surfactants.

The drag coefficients calculated by the above correlations versus bubble Reynolds number \(Re_b\) were plotted in Figure 5.10. As depicted in the figure, four different regimes can be distinguished. The “undistorted regimes” are the stokes \((0 < Re_b < 0.2)\) and viscous \((0.2 < Re_b < \text{approx.}1000)\) regimes where the drag, especially the shear drag,
is mainly determined by the liquid viscosity. When the shear stress and slip condition of the bubble surface are modified by adsorbed nanoparticles, the shear drag would be significantly enhanced. As the bubble diameter increases, the distortion and irregular motion of the bubble become pronounced and dominant for the drag force. This is the so-called “distorted regime” (approx. 1000 < Re) where a large wake due to the vortex departure is created behind the bubble. According to Ishii and Zuber (1979), in this regime the drag coefficient does not depend on the viscosity, but becomes proportional to the radius of the bubble.

Since the bubble covered with nanoparticles behaves more like a rigid sphere, the rotation-induced wake region is likely to form when the bubble Reynolds number is over 300 (Moraga et al., 1999). This means for a bubble Reynolds number 300 < Re < 1000, the bubbles may experience a drag enhancement similar to that in the distorted regime. As a result, it is expected that the transition point from the viscous regime to distorted regime may occur at a smaller Reynolds number in nanofuids, as shown in Figure 5.10.

5.1.3.5 Summary and Key Research Points

In summary, due to the adsorption of nanoparticles at bubble interfaces, the slip conditions and properties of the bubble interfaces are significantly changed. The internal circulation is suppressed, leading to an increased shear drag. Moreover, when a
bubble is covered with nanoparticles, it would partially behave like a rigid sphere and develop a rotation movement. A slanted wake could be formed behind the bubble even under comparatively low bubble Reynolds number and low Eötvös number. This slanted wake would generate a lateral Magnus force pointing towards the pipe centre and consequently make the positive-to-negative reversion of the lift force occur at a much smaller bubble diameter. Meanwhile, the slanted wake would also make bubbles in the viscous regime experience a drag force similar to that in the distorted regime, which causes the viscous-to-distorted transition point to occur at a smaller bubble Reynolds number.

However, it should be noted that the effects of nanoparticle adsorption on the interfacial forces are subject to a number of factors including the nanoparticle material, size and concentration. For example, the concentration of contaminants (e.g. nanoparticles and surfactants) can affect the progress of particle coverage considerably. This conclusion can be drawn from previously-mentioned McClure et al.’s study (2017) where a critical concentration of surfactants was observed. At low surfactant concentrations, the interface was partially covered and hence the reductions in the overall hold-up and OTR were relatively small. As the concentration of surfactants increased, the coverage of the interface also progressively increased resulting in a significantly reduced OTR until the critical surfactant concentration was reached. At this critical concentration, the gas-liquid interface was completely covered by a monolayer of surfactants. Thus above this level, additional surfactants had minimal impact. This critical concentration can be estimated as:

\[ c_{\text{monolayer}} = \frac{a M_w}{A_m N_A \rho} \times 100 \]  

(5.15)

where \( a \) is the interfacial area, \( M_w \) is the molecular weight of the antifoam, \( A_m \) is the molecular area of the surfactants, \( N_A \) is Avogadro’s number. However, McClure et al. (2017) also pointed out that it is difficult to obtain exact values for the molecular weight and area, since the surfactants are generally a blend of compounds and the exact composition of which may be proprietary. When nanoparticles exist in the liquid, the situation is more complicated and obtaining an accurate estimation of the critical nanoparticle concentration seems more difficult at current stage. All these have made formulating the interfacial forces for bubbles in nanofluids a very challenging task.

Although the adjustment to the lift coefficient and drag coefficient in this study have contributed to better results, they are, however, case sensitive and only applicable to the case of this study. In order to achieve an effective modelling and gain an in-depth understanding of the complex mechanisms, the following major focuses are recommended for the future studies:

(a) The effects of nanoparticle material, size and concentration on the structure of nanoparticle adsorption layer.

(b) The effects of nanoparticle adsorption layer on the rigidity and mobility of the bubble surface.

(c) Other possible factors like inter-particle electrostatic force, hydrophobic force and steric repulsion force.

5.1.4 Conclusions

In this study, local two-phase flow parameters including the bubble velocity and void fraction were investigated using the two-fluid model. By comparing the numerical results with the experimental data, the effects of nanoparticle adsorption at bubble interfaces on the two-phase flow behaviours were examined and the feasibility of utilizing the two-fluid model to simulate air-nanofluid bubbly flows was evaluated. Based on analyses of the numerical results, some potential mechanisms responsible for the significantly changed two-phase flow structures were also discussed and some recommendations for future work were given. The conclusions arising from this study are as follows:

(1) The spontaneous nanoparticle adsorption at bubble interfaces significantly modified the interface properties and slip conditions, which makes the bubble interface partially rigid and suppresses the inner circulation in bubbles. The rigid surface also makes the bubble develop a rotating movement and induces a wake behind a spherical bubble.

(2) The wake significantly alters the role of lift force and drag force. It is crucial to reformulate the interfacial forces when modelling nanofluid bubbly flows using the two-fluid model.
5.2 Effects of Spontaneous Nanoparticle Adsorption on the Bubble-liquid and Bubble-bubble Interaction

Abstract

Nanoparticles have been experimentally proven effective in stabilizing bubbles and enlarging the interfacial area of multi-dispersed bubbly systems. However, unlike the thorough understanding that how nanoparticles stabilize foams, the fundamental studies of the role that nanoparticles play in modifying the flow structures of bubbly flows are still very rare. This lack of mechanistic understanding and the absence of predictive theoretical models have hindered the substitution of nanoparticles for surfactants in industry. Therefore, in this study the common findings yielded from experimental and numerical investigations available in literature were analysed and summarized. It was demonstrated that the spontaneous adsorption of nanoparticles at gas-liquid interfaces is the major cause of the dramatic modification of flow structures. After analysing its influences on the bubble-liquid and bubble-bubble interactions, it was suggested that the key task when mechanistically modelling bubbly flows containing nanoparticles is to formulate the lift force, drag force, film drainage time and rupture time affected by nanoparticle adsorption.

5.2.1 Introduction

Bubbly flows, where discrete small bubbles are dispersed or suspended in liquid continuums, are widely encountered in various industries such as chemical, petroleum, mining and food processes that require large interfacial areas for efficient mixing of competing gas-liquid interactions. Maintaining the bubbly flow regime and enlarging the interfacial area have always been interests of studies with the aim to improve gas-liquid mixing during the past decades (Yao and Morel, 2004). In pursuing of larger interface area concentrations (IACs), surfactants were commonly added into the two-phase systems as they are efficient in increasing the gas-liquid interfacial area and stabilizing bubbles (Loubière and Hébrard, 2004; Rubia et al., 2010; Jia et al., 2015). In recent years, thanks to the fast advances of nanotechnology, nanoparticles have been
increasingly utilized as substitute for surfactants (Du et al., 2003) due to their unparalleled merits such as the excellent physical and chemical stabilities.

It has long been aware that nanoparticles are capable of stabilizing bubbles in quiescent liquid, such as those in foams where the volume fraction of air could be as high as 99% (Hunter et al., 2008; Worthen et al., 2013). In recent years, nanoparticles have also been found promising to stabilize dynamic multi-dispersed bubbly systems. Wang and Bao (2009) found that the bubbly-to-slug flow regime transition in a vertical tube occurred at a higher gas superficial velocity when CuO nanoparticles (0.5 wt%) were added to the nitrogen-water two-phase flow. This indicated that nanoparticles could help maintain a bubbly flow pattern with a higher void fraction than pure water. Park and Chang (2011) also experimentally investigated the two-phase flow dynamics of γ-Al2O3 nanoparticle-water mixture (0.1 vol%) and found bubbles generated through injecting air into the mixture were between 2 mm to 5 mm in diameter, which were much smaller than the bubbles in pure water (3 mm to 10 mm) injected under the same experimental conditions. The experiments also revealed that the radial void fraction distribution had a more flattened and uniform centre-peaked shape with the existence of nanoparticles in the water. The interfacial area concentration (IAC) was up to 300 m⁻³ in the nanoparticle-water mixture, almost twice as high as that in pure water.

All of these novel experimental observations have stimulated basic research on bubble hydrodynamics in nanoparticle-containing system and have called for a mechanistic understanding of the effects of nanoparticles on flow structures, which is indispensable to develop a predictive model for system design and optimization. Our primary studies (Li et al., 2016; Yuan et al., 2016), for the first time, attributed the flow structure modifications to adsorbed nanoparticles at the gas-liquid interface, which has been vastly observed in experiments (Hunter et al., 2008). The adsorption of nanoparticles was believed to affect the bubble-liquid and bubble-bubble interactions by altering the interfacial forces and bubble coalescence. Although the changes in these interactions were demonstrated to be the main cause of the smaller bubble size, uniform void fraction distribution and larger IACs observed in Park and Change’s experiments (2011) in our previous study (Yuan et al., 2016), the underlying mechanisms that how nanoparticles influence the interfacial forces and bubble coalescence process have not
been thoroughly understood. Moreover, the substitution of nanoparticles for surfactants is a pretty new technology developed in recent years and the bubbly flow containing nanoparticles is an extremely complicated physical phenomenon. A systematic review of the effects of nanoparticle adsorption on bubble dynamics and flow structures is urgently needed.

Therefore, this paper focuses on the phenomenon of nanoparticle adsorption at gas-liquid interfaces and its effects on the bubble-liquid and bubble-bubble interactions. This paper also aims to clarify the theoretical frame which in future could be used to develop predictive models for bubbly flows containing nanoparticles.

5.2.2 Nanoparticle Adsorption at Phase Interfaces

The phenomenon of nanoparticle adsorption at gas-liquid interfaces has long been recognized and vastly utilized to stabilize bubbles in liquid foams (Hunter et al., 2008). Shown in Figure 5.11 are microscopic images of nanoparticles adsorbed at the interfaces of bubbles or liquid drops submerged in another liquid. Figure 5.11(a) illustrates that MAGSILICA® H8 nanoparticles (a single-domain iron oxide core with a fully closed silica shell with a diameter of 16 ± 10 nm) suspended in liquid assembled at the surface of air bubbles submerged in an ethanol/water mixture and formed a thin layer covering the bubble (Rodrigues et al., 2011). This thin layer of adsorbed nanoparticles was also clearly observed by Lin et al. (2005) in a CdSe nanoparticle-toluene/water mixture using the Scanning Force Microscopy (SFM) in Figure 5.11(b).

Figure 5.11: (a) TEM image of air bubbles with MAGSILICA® H8 nanoparticles in ethanol/water mixture (Rodrigues et al., 2011); (b) Fluorescence confocal microscope image of the adsorbed CdSe nanoparticles at toluene/water interface (Lin et al., 2005).
Using Transmission Electron Microscopy (TEM) method, Böker et al. (2007) further demonstrated that the adsorption process include three steps: firstly, free nanoparticles diffuse to the interface; secondly, the particles pack closer and form clusters which grow to form a closely packed particle array, lowering the interfacial tension, and; finally, thermally activated exchange between adsorbed and incoming particles is observed, leading to a tightly packed layer (Figure 5.12).

Figure 5. 12: Series of TEM images of 6 nm nanoparticle adsorption to the toluene/water interface in different adsorption steps: (a) step 1; (b) step 2; (c) step 3 (Böker et al., 2007).

According to Lin et al. (2005), the adsorption of nanoparticles at the gas-liquid interface is driven by the reduction in the total interfacial free energy. The placement of a single particle with an effective radius \( r_p \) at the interface leads to a decrease of the initial interfacial energy \( E_0 \) to \( E_1 \) yielding an energy difference of \( \Delta E_1 \) (Pieranski, 1980):

\[
\Delta E_1 = \sigma r_p^2 \pi (1 \pm \cos \theta)
\]  

(5.15)

where the sign within the brackets is negative for particle removal into water (\( \theta < 90^\circ \)) and positive for particle removal into air (\( \theta > 90^\circ \)). \( \sigma \) and \( \theta \) are the surface tension and contact angle, respectively. \( \Delta E_1 \) is the so-called adsorption energy or detachment energy.

Following Equation 5.15 the energy required for a nanoparticle with diameter of 50 nm and contact angle of 80° to be detached from the water-air interface is approximately \( \Delta E_i = 65,000 \text{k}_B T \) (\( \text{k}_B \) is the Boltzmann constant and \( T \) is the absolute temperature), which is much higher than that of surfactants (generally several \( \text{k}_B T \) (Aveyard et al., 2003)). Therefore, being contrary to surfactant molecules which can dynamically adsorb to and desorb from an interface, nanoparticles can be thought of as irreversibly absorbed, which means it is almost impossible to force them out of the
interface, either by shrinkage of the bubble or thermal agitation (Rodrigues et al., 2011). As a result, this closely-packed layer of nanoparticles at the interface generates a sort of “colloidal armour” (Dickinson, 2010). This “colloidal armour”, on one hand, is found to create a steric barrier which is capable of stabilizing bubbles in liquid foams by inhibiting or even overwhelmingly stopping bubble coalescence process (Kam and Rossen, 1999; Du et al., 2003). On the other hand, the bubble surface properties and slip conditions are speculated to be significantly changed due to the presence of this “colloidal armour”. Bubbles coated with a layer of nanoparticle would deform less and be consequently more like a rigid sphere (Tomiyama et al., 1998). In addition, the part of nanoparticles immersed in the gas phase can immobilize the bubble surface and change the slip condition from free-slip to no-slip, resulting in the partially or completely supressed inner circulation flow (Dijkhuizen et al., 2010a). Since bubble-liquid and bubble-bubble interactions which control the bubble’s movements, distribution and size, are predominantly influenced by the bubble surface properties and bubble coalescence process, it is crucial to clarify the mechanisms of the effects of nanoparticles on these two interactions.

5.2.3 The Influences of Nanoparticles on Bubble-liquid Interactions

5.2.3.1 Bubble-liquid Interaction

Hydrodynamic interactions between the gas and liquid phases are responsible for the complexity of gas-liquid flows. Interfacial forces are almost always the dominant components of these interactions and their formulations are critical to the prediction of gas-liquid flows. Forces exerted on a bubble moving in continuous liquid include drag force $\vec{F}_D$, lateral lift force $\vec{F}_L$, wall lubrication force $\vec{F}_W$ and turbulent dispersion force $\vec{F}_{TD}$. The total interfacial force $\vec{F}_{Ig}$ on the bubble is:

$$\vec{F}_k = \vec{F}_D + \vec{F}_L + \vec{F}_W + \vec{F}_{TD} \quad (5.16)$$

$$\vec{F}_D = \frac{3}{4} \frac{C_D}{d_s} \alpha_g \rho_i |\vec{U}_g - \vec{U}_i| (\vec{U}_g - \vec{U}_i) \quad (5.17)$$

$$\vec{F}_L = C_L \alpha_g \rho_i (\vec{U}_g - \vec{U}_i) \times (\nabla \times \vec{U}_i) \quad (5.18)$$
\[
\bar{F}_w = - \frac{\alpha_s \rho_l (U_g - U_i)}{d_b} \max \left(0, C_{w1} + C_{w2} \frac{d_b}{y_w} \right) \hat{n} \tag{5.19}
\]
\[
\bar{F}_{TD} = -C_{TD} \rho \kappa_i \nabla \alpha_i \tag{5.20}
\]

where \(C_D, C_L, C_{W1}\), and \(C_{W2}\), and \(C_{TD}\) denote drag coefficient, lift coefficient, wall lubrication coefficients and turbulent dispersion coefficient, respectively. The formulation of these coefficients has been strongly empirical due to the extreme complexity. Although dozens of correlations have been proposed for these coefficients, considerable uncertainties and discrepancies remain being reported due to their empirical nature. It is worth noting that these interfacial forces are all closely related to liquid velocity filed surrounding the bubble (Kolev, 2012). An insight into the liquid flow around a nanoparticle-covered bubble is thus needed.

When a spherical gas bubble having a clean interface, moves at a constant velocity \(U\) through a continuous liquid phase, its streamlines are open, and in particular there are no wakes behind (Figure 5.13(a)) (Brenner, 2013) because no shear exists on the bubble interface. As the Weber number \((\text{We}=\rho U^2 r_b/\sigma)\) increases, the inertial distorts the bubble from spherical to oblate-ellipsoidal and spherical cap shapes. When the distortion is significant, flow separation and wake occur at the back end if the bubble Reynolds number \(Re_b\) is larger than 125 (Ryskin and Leal, 1984) (Figure 5.13(b)). However, for solid spheres as long as the Reynolds number is larger than about 12, flow separation and wake formation can always occur (Clift et al., 1978; Johnson and Patel, 1999). This fact suggests that the reduction in a spherical bubble’s interfacial mobility can cause a wake to form at its back end (Fdhila and Duineveld, 1996; Mclaughlin, 1996; Wang et al., 2002). When it comes to nanoparticle-containing system, due to the so-called “colloidal armour” of nanoparticles, bubbles will be partially rigid and immobile, and become more resistant to deform (Sugiyama et al., 2001). A wake region could probably form behind the nanoparticle-covered bubble (Wang et al., 2002; Fukuta et al., 2008). Meanwhile, under the action of liquid velocity gradient and fluid shear, the nanoparticle-covered bubble tends to develop a rotating movement (Kurose and Komori, 1999). This rotating movement has been demonstrated to induce wake asymmetries, as illustrated in Figure 5.13(c) (Taneda, 1957).
Figure 5.13: Flow field surrounding the bubble: (a) spherical bubbles in pure liquid; (b) distorted bubbles in pure liquid; (c) spherical bubbles in nanoparticle-containing system.

5.2.3.2 The Lift Force

Lift force generally acts in the direction normal to the relative motion of fluid and bubbles, and largely controls the transverse motion of bubbles in a vertical flow. For small spherical bubbles in pure liquid shear flow, a lateral force is caused by the pressure difference due to a liquid velocity gradient (Figure 5.14(a)). This lateral force is the so-called shear-induced lift force, which acts towards the descending liquid velocity gradient, or in another word, towards the pipe wall for a spherical bubble rising in an upward liquid flow ($U_g > U_l$). The lift coefficient $C_L$ is thus positive with a value ranging from 0.25 to 0.5 depending on the bubble Reynolds number and liquid viscosity (Zun, 1980; Auton, 1987; Lance and de Bertodano, 1994). For distorted bubbles in pure liquid, besides the shear-induced lift force, another lateral force arises due to the complex interactions between the bubble wake and the liquid shear field.
(Serizawa and Kataoka, 1994). According to Tomiyama et al. (1995) this wake-induced lift force acts in an opposite direction of the shear-induced lift force and causes a direction reversal when the wake becomes strong enough (Figure 5.14(b)).

Figure 5. 14: The lift force acting on: (a) spherical bubbles in pure liquid; (b) distorted bubbles in pure liquid.

Tomiyama et al. (2002) developed an empirical $C_L$ correlation which has allowed modelling the transverse migration of spherical and distorted bubbles in pure liquid:

$$
C_L = \begin{cases} 
\min \left[ 0.288, \tanh(0.121R_e, f(Eo^*)) \right] & Eo^* \leq 4 \\
0.00105Eo^{*2} - 0.0159Eo^{*3} - 0.0204Eo^{*4} + 0.474 & 4 < Eo^* \leq 10 \\
-0.27 & 10 < Eo^* 
\end{cases} 
$$

(5.21)
where $Eo^*$ is the modified Eötvös number based on the maximum bubble horizontal dimension $d_H$. The Tomiyama lift coefficient is plotted against the bubble size in Figure 5.15. For air bubbles rising in pure water, the negative-to-positive transition occurs at a critical bubble diameter of $d_c=5.8$ mm (Liu, 1993; Grossetete, 1995; Sakaguchi et al., 1996).

When bubbles are coated with nanoparticles, they are more likely to behave like rigid spheres rather than deformable bubbles due to the increased rigidity and restricted mobility. As for the rigid sphere, both Kurose and Komori (1999) and Bagchi and Balachandar (2002) showed that the lift coefficient $C_L$ decreases with increasing bubble Reynolds number and takes near-zero value at $Re_b=100$. Beyond this value, $C_L$ keeps slightly decreasing and it takes the small negative value, indicating that the lift force on a rigid sphere acts in the opposite direction of that on a free-slipping bubble. In fact, similar findings have been obtained in studies of bubbles contaminated with surfactants (Fukuta et al., 2008; Dijkhuizen et al., 2010). Our previous studies (Li et al., 2016; Yuan et al., 2016) also revealed that $C_L$ can be negative for small spherical nanoparticle-coated bubbles.

The Tomiyama correlation (Equation 5.21) (Tomiyama et al., 2002) was incorporated in the two-fluid model employed in our previous study (Li et al., 2016) to simulate air-water bubbly flows with nanoparticles. The numerical results were then compared against the experimental data from Park and Chang (2011). In the computations a wall-peaked void fraction distribution was yielded despite the factual centre-peaked distribution observed in the experiments (Figure 5.16(a)). The reason that led to this difference was found to be the positive lift coefficient with a value of 0.288 obtained by Equation 5.21 where the employed bubble diameter was 3 mm on average according to the experimental measurements. When a negative value $C_L=-0.025$ was used in the simulation, a good agreement with the experimental data was achieved (Figure 5.16(b)).
Figure 5.16: Comparison of predicted flow parameters against experimental data of bubbly flows containing nanoparticles with: (a) Tomiyama model (Equation 5.21); (b) $C_L = -0.025$ (Yuan et al., 2017).

Since the void fraction distribution reflects the bubble distribution, this result indicates that the lift coefficient for small spherical bubbles in nanoparticle-containing system can be negative and under the action of which these bubbles migrated towards the pipe centre. The widely accepted Tomiyama correlation (Equation 5.21) is thus not feasible to nanoparticle-covered bubbles. The positive-to-negative transition of the lift coefficient occurs at a much smaller critical bubble diameter, as shown in Figure 5.15. In order to develop a model appropriate for the lift force in nanoparticle-containing system, two plausible mechanisms that how nanoparticles reverse the direction of the lift force are analysed in this study.
Figure 5. 17: Contributions of pressure $C_{L,p}$ and viscous stress $C_{L,v}$ to the total lift coefficient acting on: (a) a contaminated bubble (Fukuta, Takagi et al., 2008); (b) a rigid sphere (Kurose and Komori, 1999).

**Marangoni effect**

The lift force acting on a surfactant-contaminated bubble in a linear shear flow was numerically studied by Fukuta et al. (2008). They found the lift force decreased from the positive value of a clean bubble to a negative value, when the bubble gradually became fully contaminated. For the first time, they related this reduction to a nonaxisymmetric distribution of pressure on the bubble surface which was caused by the Marangoni effect. As explained by Fukuta et al. (2008), a surface concentration
distribution exists along the bubble surface because the surfactant is swept off the rear part and accumulates in the rear part as the bubble rises. Due to this surfactant accumulation in the rear part, a variation of surface tension along the surface is developed and this causes a tangential shear stress on the bubble surface. This is known as Marangoni effect and the tangential shear stress is the so-called Marangoni stress. Since nanoparticles act in many ways like surfactants (Binks, 2002) and tend to accumulate in the rear part of a rising bubble, it is reasonable to extrapolate this Marangoni effect may affect the lift force acting on a nanoparticle-coated bubble in a similar way. Due to this Marangoni stress, both pressure and viscous stress on the bubble surface can become asymmetrically distributed. The lift coefficients due to pressure $C_{L,p}$ and due to viscous stress $C_{L,v}(C_L = C_{L,p} + C_{L,v})$ are thus inevitably changed. Figure 5.17(a) illustrates that with more surfactants adsorbed on bubble surface (corresponding to an increase of Langmuir number), the pressure contribution $C_{L,p}$ decreases dramatically to a negative value. When the bubble is fully coated (maximum Langmuir number), the viscous stress contribution $C_{L,v}$ becomes dominant, giving the negative value of the total lift coefficient $C_L$.

In fact, when a bubble is fully covered with nanoparticles and behaves like a rigid sphere, the lift force acting on this bubble is also influenced by the Reynolds number $Re_b$ (Kurose and Komori, 1999). Shown in Figure 5.17(b) are the contributions of pressure $C_{L,p}$ and viscous stress $C_{L,v}$ acting on a rigid sphere in a homogeneous linear shear flow for fluid shear rate $\alpha^* = 0.2$. Both coefficients $C_{L,p}$ and $C_{L,v}$ change their signs from positive to negative in the range $1 \leq Re_b \leq 100$. According to Park and Chang’s experiment (2011), the bubble Reynolds number in the bubbly flow was estimated to be 1000. Obviously, the lift coefficient will be negative even for small spherical nanoparticle-coated bubbles.

**Wake effect**

As shown in Figure 5.13(c), a slanted wake region induced by the immobile surface and rotating movement can be found behind the nanoparticle-coated bubble. Since the size of the wake is generally of the same order as that of the bubble itself, its effect on body forces cannot be neglected. In the wake region, when a vortex is shed, the space it
occupied behind the bubble is replenished by liquid moving more slowly than the rotational velocity of the vortex (Moraga et al., 1999). A significant velocity reduction occurs due to the sharp turn made by the incoming fluid to occupy the volume immediately after the body. As a result, an increase in pressure will be generated by the decrease in the velocity of the fluid. Therefore, when a vortex is shed, a transient lateral force on the bubble will arise (Jordan and Fromm, 1972; Alajbegović et al., 1998; Moraga et al., 1999). Sakamoto and Haniu (1995) discovered that vortices at the higher relative velocity side always grow faster and larger than those at the lower relative velocity side. Then the smaller vortices will be engulfed by the larger ones before they form a separate vortex and detach. In the absence of shedding, the lateral force is thus always toward the lower relative velocity side, which is opposite to the direction of the shear-induced lift for a rising bubble in an upward flow. The total lift force on a nanoparticle-coated bubble is thus expected to be a consequence of two competing factors: shear and wake effects. The total lift coefficient $C_L$ is given by the sum of shear-induced lift coefficient $C_{LS}$ and wake-induced lift coefficient $C_{LW}$:

$$C_L = C_{LS} + C_{LW} \quad (5.22)$$

Combining the experimental data with numerical data, the total lift coefficient in turbulent shear flows was correlated by Moraga et al. (1999) in terms of both bubble Reynolds number $Re_b$ and vorticity Reynolds number $Re_ω$:

$$C_L = 0.17 \exp\left(\frac{Re_b Re_ω}{4.2 \times 10^3}\right) \quad (5.23)$$

According to Equation 5.23, no wake-induced lift force is expected for Reynolds numbers below 300 and consequently shear effect should be dominant. As the Reynolds number increases, wake effect becomes increasingly important and eventually reverses the sign of the lift coefficient to negative (Figure 5.18).

Actually, no matter Equation 5.21 or Equation 5.23 are both empirical correlations. As pointed out by Moraga et al. (1999), an accurate determination of the magnitude of the lift force induced by the wake effect is still very difficult, the main problems being the complexity of the wake structure and its elusiveness to an analytical treatment. Therefore, more fundamental and analytical studies are still urgently needed in future.
Figure 5. 18: The lift force acting on spherical bubbles in nanoparticle-containing system.

5.2.3.3 The Drag Force

The drag force is one of the most important forces encountered in bubbly flows, and it dominantly controls the rise velocity of the bubbles in a vertical flow. It is a result of the shear and form drag, which are due to viscous surface shear stress and pressure distribution around the bubble, respectively. According to Ishii and Zuber (1979), when calculating the drag coefficient $C_D$ in Equation 5.17, the bubbly flow behaviours of pure liquid were categorized into four different regimes: stokes, viscous, distorted and churn. The stokes ($0 < Re_b < 0.2$) and viscous ($0.2 < Re_b < 1000$) regimes are characterized by the “undistorted particles” where the distortions of the bubbles are negligible and the drag coefficient $C_D$ mainly depends on the bubble velocity and liquid viscosity. As the bubble diameter increases, the shape of the bubble is gradually changed from spherical to oblate-ellipsoidal and then spherical cap. A vortex system will develop behind the bubble, where the vortex departure creates a large wake region. This process happens in the distorted and churn regimes which are known in literature as “distorted particle” regimes ($1000 < Re_b$). In these two regimes, the distortion and irregular motions become pronounced and the drag coefficient $C_D$ becomes proportional to the bubble radius and Reynolds number. Thus a mixture viscosity model was developed by Ishii and Zuber (1979) to obtain each drag coefficient correlations for the individual flow regimes. The drag coefficient as calculated is plotted in terms of the bubble Reynolds number in Figure 5.19.
Besides the influences of aforementioned bubble’s radius and Reynolds number, the surface properties and slip condition also play important roles. It has been well known the drag coefficient of a solid particle can be almost three times large of the corresponding drag coefficient of the bubble with the same radius and Reynolds number. When a clean bubble is contaminated with impurities, these impurities such as surfactants and nanoparticles can bridge the gap existing between the behaviour of a clean bubble and a solid particle by immobilizing (at least partly) the bubble surface (Harper, 1972; Clift et al., 1978; McClure et al., 2014). As a result, the drag force on a contaminated bubble increases from that of a clean bubble to that of a rigid sphere (Cuenot et al., 1997). In addition, Tomiyama et al. (1998) believed the aggregation of impurities can also increase the shear drag by inducing the no-slip condition and hindering the internal circulation within the bubble (Figure 5.20). With the
consideration of this effect, an empirical correlation was proposed to account for the drag enhancement (Equation 5.25).

\[ C_{D,Tomiyama} = \max \left[ 24(1 + 0.15 \frac{Re_b}{Re_b^{0.687}}) / Re_b, 8Eo / 3(Eo + 4) \right] \] (5.25)

Figure 5. 20: The effect of contaminants: (a) ultra-pure liquid with free-slip boundary condition; (b) slightly contaminated liquid with a limited circulation inside the bubble; (c) fully contaminated bubble with no-slip boundary condition (Dijkhuizen et al., 2010a).

Recently, McClure et al. (2015) improved the classic Grace model by multiplying an empirical constant \( k_s = 1.6 \sim 2.2 \) to include the effects of adsorbed surfactants on the drag enhancement (McClure et al., 2014):

\[ C_D^s = k_s C_{D,Grace} f(\alpha) \] (5.26)

As nanoparticles are found to behave like general surfactants in many ways such as adsorbing at the bubble interfaces and changing the slip condition (Binks, 2002), it is extrapolated that adsorbed nanoparticles might play a similar role in increasing the shear drag. Following the modified Grace model, our previous study (Yuan et al., 2016) introduced the same empirical constant \( k_s \) to the Ishii-Zuber model (Ishii and Zuber, 1979) (Equation 5.24) to account for the influence of nanoparticles and further expanded its range to \( k_s = 1.6 \sim 3.0 \):

\[ C_D^s = k_s C_{D,Ishii} \] (5.27)

The numerical results of bubble velocity obtained with the above-mentioned models showed that accounting for the influence of adsorbed nanoparticles on shear
drag through a simple coefficient only has very limited impact on the predicted value (Figure 5.21). There must be other factors that need to be considered.

![Figure 5.21: Comparison of predicted bubble velocity against experimental data of bubbly flows containing nanoparticles with different drag models (Yuan et al., 2017).](image)

**Marangoni effect**

Fukuta et al. (2008) found that the Marangoni effect induced by the accumulation of surfactants on bubble surface not only influences the lift force but also increases the drag force. As aforementioned, when surfactants adsorb on the bubble surface, a tangential shear stress can develop. This implies that a shear-free boundary condition is no longer imposed in the liquid at the gas-liquid interface, and this leads to an increase in the drag force. Duineveld (1994) and Bel Fdhila and Duineveld (1996) carried out experiments with bubbles rising in water contaminated with surfactants. Below a critical bulk concentration, they found that the final rise velocity is insensitive to the presence of surfactants, whereas the rise velocity decreases abruptly to the value corresponding to a solid sphere above the critical bulk concentration. Much effort has been devoted to modelling the phenomena reported above. The most widely employed one is the stagnant-cap model, where the bubble surface is divided into two different regions separated by a stagnant-cap angle $\theta_c$ (Savic, 1953). For $\theta_s < \theta_c$ ($\theta_s$ is an angle from the front stagnant point), the surfactant surface concentration $\Gamma$ is zero and the liquid remains free to slip along the interface; whereas in the rear of the bubble ($\theta_s > \theta_c$),
Γ is nonzero and the relative velocity of the fluid along the interface $u_s$ vanishes. The drag coefficient for a surfactant-contaminated bubble can be calculated by the correlation proposed by Sadhal and Johnson (2006):

$$\frac{C_D(\theta_s) - C_D(\pi)}{C_D(\theta_s) - C_D(\pi)} = \frac{1}{2\pi} \left[ 2(\pi - \theta_s) + \sin \theta_s + \sin 2\theta_s - \frac{1}{3} \sin 3\theta_s \right]$$

(5.28)

As aforementioned, Marangoni effect can also be found on nanoparticle-coated bubbles. The stagnant-cap model is probably capable of describing the distribution of nanoparticles at bubble surfaces. Thus employing Equation 5.28 might be a potential way to calculate the drag coefficient for a nanoparticle-containing system.

**Wake effect**

When the boundary condition around $\theta_s = \theta_c$ abruptly changes from a shear-free to a no-slip condition, a marked peak in the interfacial vorticity is produced. Thus there is more vorticity injected in the flow than in the case of a uniform no-slip condition, and this results in a larger wake in length (Cuenot et al., 1997) and volume (McLaughlin, 1996) of surfactant-contaminated bubbles than those of solid spheres moving at the same Reynolds number. Moreover, the wake effect becomes much stronger when the bubble Reynolds number is over 300 (Moraga et al., 1999) and causes distortion and irregular motion to the bubbles. The contaminated bubbles may consequently experience a drag enhancement similar to that in the distorted regime when the bubbly Reynolds number is in the range of $300 < Re_b < 1000$. As a result, it is expected that the transition point from the viscous regime to distorted regime may occur at a smaller Reynolds number in nanoparticle-containing system, as shown in Figure 5.22.
Figure 5. 22: The predicted drag coefficient as a function of bubble Reynolds number with different drag models (Yuan et al., 2017).

However, due to the lack of experimental data, taking the Marangoni and wake effects into account to model the drag force is still a challenge. For the stagnant-cap model, the major difficulty is the determination of the cap angle $\theta_c$ as a function of ambient nanoparticle surface concentration. Since the nanoparticles are irreversibly absorbed, which is contrary to surfactant molecules that can dynamically adsorb to and desorb from the surface, how to emphasize this difference and substitute a suitable cap angle still remains a difficult problem.

5.2.4 The Influences of Nanoparticles on Bubble-bubble Interactions

5.2.4.1 Bubble-bubble Interaction

In gas-liquid flows, the effects of coalescence and break-up through the interactions among bubbles have attracted considerable attention, since they largely influence the temporal and spatial evolution of the two-phase structure by deciding the bubble size. Compared to break-up, coalescence was demonstrated dominant in the case of upward bubbly flows in small-diameter vertical tubes (Li et al., 2010). In view of this, only bubble coalescence is considered in this study. According to the film drainage model proposed by Shinnar and Church (1960), bubble coalescence occurs within three steps: contact, thinning and rupture. Firstly, two bubbles come into contact with each other in
the liquid phase, flattening the bubble surfaces against each other and trapping a thin liquid film between them. The initial thickness $h_0$ of this film is typically $10^{-4}$ m (Kirkpatrick and Lockett, 1974). The first step is controlled by the hydrodynamics of the bulk liquid phase. Secondly, this intervening liquid film thins to a critical thickness $h_f$ (usually estimated as $10^{-8}$ m (Kim and Lee, 1987)) before it ruptures. If this thinning process takes longer than the bubble contact time, coalescence will not occur. The second step is controlled by the hydrodynamics of the liquid film. Thirdly, once the film is sufficiently thin it will rupture via an instability mechanism. This step is very rapid in comparison to the first two and it is usually not counted in the coalescence time.

Figure 5. 23: Schematic overview of the coalescence process of two bubbles.

According to the film drainage model, not all collisions lead to coalescence. The concept of collision efficiency $\lambda$ is introduced to account for the probability of bubble coalescence:

$$\lambda = \exp\left(-\frac{t_{dr}}{\tau}\right)$$  \hspace{1cm} (5.29)

A larger collision efficiency leads to a larger mean bubble diameter and vice versa. In Park and Chang’s experiment (2011), the measured bubble diameters were between 2 mm to 5 mm in air-water bubbly flows with nanoparticles, which were much smaller than those (3 mm to 10 mm) without nanoparticles under the exactly same bubble injection condition. Since only coalescence mechanism is considered in this study, it is reasonable to extrapolate the decrease of bubble size in nanoparticle-containing system is probably due to a reduced coalescence efficiency.

According to Equation 5.29, the coalescence efficiency is determined by the contact time $\tau$ and the drainage time $t_{dr}$ (Coulaloglou, 1975). The contact time $\tau$ is
controlled by the external liquid flow and turbulence in the bulk (Chesters, 1991). Since
the concentration of nanoparticles in the bulk flow filed is as low as 0.1 vol% in the
Park and Chang’s experiments, the nanoparticle-water mixture could be assumed at
near-molecular level and treated as a pseudo-homogenous liquid. Therefore, the
nanoparticles would probably not influence the contact time. The drainage time \( t_{dr} \)
which is the time required for the thinning process, is determined by the internal liquid
flow in the intervening film between the bubbles. Du et al. (2003) experimentally
investigated the stability of bubbles coated with silica particles (primary diameter of 20
nm) and concluded that the adsorbed nanoparticles hindered the water flow at bubble
surface and slowed down film thinning process. Thus the drainage time of the liquid
film in nanoparticle-containing systems might be elongated. Actually this hypothesis is
consistent with the simulation results in our previous parametric study (Yuan et al.,
2016). In this study a correction coefficient \( k_d \) ranging from 1.0 to 2.0 was added to the
widely used Prince and Blanch (1990) model to calculate the drainage time.

\[
t'_{dr} = k_d t_{dr}
\]  
\[
t_{dr} = \left( \frac{r_b^3 \rho_f}{16 \sigma} \right)^{1/2} \ln \frac{h_0}{h_f}
\]

When the correction coefficient took the value of \( k_d = 1.02 \), the model achieved
closer predictions of void fraction with the experiment data, as shown in Figure 5.24.
This indicates that the drainage time is indeed elongated by nanoparticles. With the
purpose of further comparison, the predicted bubble size distribution was depicted in
Figure 5.25. When the coefficient \( k_d \) increased from 1.0 to 2.0, the largest proportion of
bubbles moved from group 4 (4.5~6 mm) to group 2 (1.5~3.0 mm), which
demonstrated the inverse relationship between the drainage time and bubble diameters.

Although better agreement has been achieved by employing Equation 5.30 in our
previous study, the correction coefficient \( k_d \) is case sensitive and subject to a number of
factors. In order to develop a mechanistic model, the underlying mechanisms that how
nanoparticles elongate the drainage time have to be thoroughly understood.
Figure 5. 24: Comparison of predicted void fraction against experimental data of the bubbly flows containing nanoparticles (Yuan et al., 2016).

![Graph showing void fraction vs. r/R for different values of k_d.]

Figure 5. 25: Comparison of predicted bubble size fraction when k_d takes the value of k_d=1.0-2.0 (Yuan et al., 2016).

### 5.2.4.2 Thinning Process

As aforementioned the drainage time is determined by the thinning of the liquid film. According to Oolman and Blanch (1986), this thinning process of a clean liquid film in pure liquid is predominantly driven by the capillary pressure induced by the variations in the curvature of gas-liquid interface. The interface is very close to flat at the centre of the film and the pressure at that point equals to the pressure inside the bubble.
Outside the film a surface tension force towards the centre of the bubble is induced by the curvature of the bubble’s surface. And this surface tension force has to be balanced by a change in pressure across the interface. Thus the pressure in the bulk liquid outside the film is smaller than the pressure at the film’s centre. This pressure difference (Equation 5.32) is the so-called capillary pressure, pushing the liquid in the film to flow outside (Figure 5.26):

\[ \Pi_c = \frac{2\sigma}{r_b} \]  

(5.32)

![Diagram](image)

Figure 5. 26: Drainage of a liquid film under capillary pressure (Rio and Biance, 2014).

The thinning process could be governed using the conservation equations of mass and momentum (Equation 5.33&34).

\[ \frac{\partial h}{\partial t} + \frac{\partial hu_x}{\partial x} = 0 \]  

(5.33)

\[ \rho \frac{\partial u_x}{\partial t} = \eta \Delta u_x - \nabla \Pi + \rho g \]  

(5.34)

Where \( h \) is the film thickness, \( u_x \) is the liquid velocity, and \( \Pi \) is the pressure gradient. These two equations must be closed using appropriate boundary conditions at the gas-liquid interface, which is crucial to determine properly the drainage dynamics. Mysels (1959) investigated the drainage of a foam film and proposed two limiting cases of drainage, depending on the mobility of interfaces: zero stress at a mobile interface and zero velocity at an immobile interface. Rio and Biance (2014) compared the results obtained with the mobile (Howell and Stone, 2005) and immobile (Aradian et al., 2001) boundary conditions and found that it takes almost 80 \( \mu \)s for the immobile film to reach
$10^8$ m from $10^6$ m, whereas it needs only 0.7 $\mu$s in the mobile case. This indicates that the immobility of the interface can significantly increase the film drainage time.

**Surface mobility and rigidity**

A number of experimental and numerical studies have demonstrated the adsorbed nanoparticles can restrict the mobility of bubble interface. Lin and Slattery (1982) developed a theoretical model for the thinning of the liquid film which forms as a bubble approaches an interface. They found that very small surface tension gradients are sufficient to immobilize the interface. Worthen et al. (2013) further suggested the addition of nanoparticles could increase the effective viscosity of the injected gas in the liquid and thereby reduce the bubble mobility. It is thus reasonable to extrapolate that restricting the mobility of the bubble surface through nanoparticle adsorption might be one of the possible mechanisms responsible for the elongated drainage time.

With the consideration of this effect, the Equation 5.31, which was proposed by Prince and Blanch under the assumption that the bubble surface is fully mobile and zero-stress (Figure 5.27(a)), is no longer feasible in nanoparticle-containing system. When nanoparticles gradually assemble at the interface and partially cover the bubble (Figure 5.27(b)), the liquid flow becomes quasi-steady creeping. Chesters (1991) defined the drainage time for partially mobile interfaces:

$$I_{dr} = \frac{\pi \mu F^{1/2}}{2(2\pi \sigma) r_b^{3/2}} \left( \frac{1}{h_f} - \frac{1}{h_b} \right)$$

(5.35)

Figure 5. 27: The velocity profile of the liquid in the film with: (a) fully mobile interfaces; (b) partially mobile interface; (c) fully immobile interfaces (Liao and Lucas, 2010).
When bubbles are fully covered with nanoparticles, their surfaces become fully immobile. According to Marrucci (1969), the viscous effects, instead of the inertial effects, dominantly controlled the film thinning process. The liquid is expelled from between these immobile surfaces by a laminar flow. As illustrated in Figure 5.27(c), the velocity profile in the film becomes parabolic with no slip at the surface. Considering the fully restricted mobility, Chesters (1991) derived the drainage time:

\[
I_{dr} = \frac{3\mu FL}{16\pi\sigma^2} r_b^2 \left(\frac{1}{h_f} - \frac{1}{h_0^2}\right)
\]

(5.36)

Since the total film drainage time is predicted to be tens and even hundreds times longer for immobile surfaces than mobiles surfaces, prediction of the transition from very rapid to very slow coalescence becomes an important issue. Marrucci (1969) related this transition to the particle concentration c and proposed a model for the critical concentration:

\[
c_c = 0.084 R T (\sigma A_{Ht}^2 / r_b^2)^{1/3} \left(\frac{\partial \sigma}{\partial c}\right)^{-2}
\]

(5.37)

where \(A_H\) is the Hamaker constant and \(R^*\) is the ideal gas constant. The above analysis is based on the parallel model which assumes that the surfaces of coalescing bubbles deform into two parallel discs (Figure 5.28(a)). Actually, when nanoparticles fully cover the bubble, their surfaces can be slightly deformed and behave as nearly rigid spherical particles (Figure 5.28(b)). For two non-deformable spheres, the drainage time is defined as (Chesters, 1991):

\[
I_{dr} = \frac{3\pi \mu L r_b^2 \ln h_0}{2F r_b} \ln \frac{h_0}{h_f}
\]

(5.38)

However, it should be noted that the assumption of a non-deformable figure is only reasonable for small bubbles. In most applications where large bubbles exist, the deformation of bubble surface during the collision has to be considered even with contaminants like nanoparticles. Therefore, the aforementioned parallel model is still feasible for nanoparticle-coated bubbles.
Another factor influencing the thinning process is the pressing force that brings two bubbles to coalescence. This pressing force is usually described as the capillary pressure between the bubbles and the inter-film fluid. When nanoparticles with a zero contact angle that are completely resting in the liquid film, lie between the two bubbles (Figure 5.29), the capillary pressure is changed.

As drainage occurs, the bubbles form a meniscus around the particle. The curvature of the meniscus induces a net surface tension force towards the centre of the bubble which has to be balanced by a change in pressure. The pressure at the centre of the film with nanoparticles is no longer equal to the pressure inside the bubble but becomes much smaller. As a result, the capillary pressure $\Pi_C$ for bubbles with
nanoparticles is much smaller than the capillary pressure $\Pi_c$ for bubbles without nanoparticles. $\Pi_c^*$ can be expressed as:

$$\Pi_c^* = \frac{2\sigma}{r_b} \left(1 - \frac{r_c}{r_m}\right)$$

(5.39)

where $r_m$ is the curvature radius of the meniscus. With a smaller capillary pressure, a slower thinning process will happen, which leads to a longer drainage time. As shown in Equation 5.39, not only the curvature of the meniscus can affect the capillary pressure, but the surface tension $\sigma$ plays an important role. In a number of studies, nanoparticles have been demonstrated to be effective in lowering the surface tension of interfaces (Böker et al., 2007). During the thinning process, the surface area increases whereas the surface concentration of adsorbed nanoparticles decreases. Since surface tension is an inverse function of nanoparticles’ concentration, a surface tension gradient can develop along the bubble surface. According to Oolman and Blanch (1986), the change of surface tension $\sigma$ due to the existence of impurities like nanoparticles can be expressed as:

$$\Delta\sigma = \frac{1}{h} \left(\frac{2c}{RT}\right)(\frac{\partial\sigma}{\partial c})^2$$

(5.40)

It is thus important to take the change of surface tension into account when calculating the capillary pressure in nanoparticle-containing system.

For pure liquid, it is true that the capillary pressure is the only pressure acting on the liquid film. But Oolman and Blanch (1986) found that when a second component exists in the liquid, other pressures resisting the film thinning can develop. These pressures include the electrostatic double layer force and steric repulsion force. Langevin (2015) pointed out that these disjoining pressures are mainly responsible for stabilizing foams after conducting a mechanistic analysis.

In fact, for nanoparticles with ionisable surface groups (e.g. latex or silica), the part of the particle immersed in the aqueous phase will become charged (Figure 5.30). Thus an electrostatic double layer can be established. Sagert and Quinn (1978) investigated the effect of electrostatic forces on thinning process and they believed that this repulsive force can balance the capillary pressure and cause the film thinning to
stop at an equilibrium film thickness. The equation of electrostatic double layer force is given by (Bhattacharjee et al., 1998):

\[
\Pi_s = (64\pi k_B T \rho_s \gamma^2 / \kappa^2) \exp(-\kappa h)
\]

where \( \rho_s, \gamma, \) and \( \kappa \) represent the density of electric charge in the bulk solution, reduced surface potential and Debye screening length, respectively.

\[
\Pi_s = \frac{k_B T}{s^3} \left[ \frac{2L}{\delta} \right]^{9/4} - \left( \frac{\delta}{2L} \right)^{3/4}
\]

where \( \delta, L \) and \( s \) represent the separation between the surfaces, the thickness of the polymer layer, the mean distance between the attachment points. As the drainage occurs, the above-mentioned disjoining pressures withstand the capillary pressure. The

Figure 5. 30: Electrostatic double layer force between two nanoparticle-adsorbed bubble interfaces.

In addition to electrostatic force, Samanta and Ghosh (2011) believed that the reduced bubble coalescence in contaminated systems is mainly due to the steric force imparted by the adsorption of amphiphilic contaminants at gas-liquid interfaces. The adsorbed layer encounters a reduction in entropy when confined in a very small space as the bubble approaches to each other. Since the reduction in entropy is thermodynamically unfavourable, their approach is thus inhibited. According to Böker et al. (2007), some nanoparticles such as Janus-particles like polymers have two surface regions: polar surface region and apolar surface region. These nanoparticles are surface active and amphiphilic (Böker et al., 2007). It is reasonable to extrapolate that when two bubbles approach to each other, similarly to the polymeric surfactant, the hydrated head groups of adsorbed nanoparticles will be overlapped, generating a steric repulsion force. This force could be calculated by (Samanta and Ghosh, 2011):
pressure gradient $\Pi$ in Equation 5.34 thus decreases, slowing down the liquid flow in the film and elongating the drainage time.

Langevin (2015) further found that with the thinning of the liquid film, the disjoining pressures can also affect the equilibrium thickness of the liquid film. In the absence of nanoparticles and the induced disjoining pressures, the capillary pressure drains the liquid film to the critical thickness $h_f$ and film surface waves rupture the film rapidly. Therefore, in pure liquid the film rupture time is much smaller than the drainage time and usually not counted in the coalescence time. With the existence of nanoparticles, the disjoining pressure can equilibrate the capillary pressure at a thickness larger than the critical thickness $h_f$. When this happens, the rupture of the film might become not that rapid and the rupture time even can be comparable to the drainage time. If the coalescence time, which includes the drainage time and rupture time, is longer than the bubble contact time, coalescence will still not occur. Therefore, the effects of nanoparticles on the rupture process have to be fully understood as well.

### 5.2.4.3 Rupture Process

It has been proposed in the literature that the growth of thermodynamic instability of the liquid film is the main factor that leads to the film rupture (Vrij, 1964). These instabilities are caused by the thermal fluctuations which can corrugate a deformable interface. Initial amplitude of surface wave at a single interface is very small, approximately $10^{-10} - 5 \times 10^{-10}$ m (Valkovska et al., 2002). While thinning, the amplitude of the surface waves keeps growing. Once the wave amplitude reaches to the critical film thickness $h_f$, the film will rupture and the two bubbles start to coalesce.

Thermal corrugations of the interface of thin liquid films were first observed through light-scattering experiments (Figure 5.31(a)) (Vrij, 1964). These fluctuations are inhibited by surface tension but enhanced by Van der Waals attractive interactions between both sides of the film. Then taking into account both effects, Vrij and Overbeek (1968) determined the critical wavelength $\Lambda_c$ of the thermal fluctuations that are amplified by the follow expression:

$$\Lambda_c = 2\pi \sqrt{\frac{\pi \sigma h^4}{\Lambda_H}}$$  \hspace{1cm} (5.43)
The rupture time, which is the time required for a surface wave to develop to the critical film thickness $h_f$, is calculated as:

$$t_{rp} = 10 \times 96 \pi^2 \mu \sigma A_H^{-2} h_f^5$$

(5.44)

where $t_{rp}$ is estimated to be 330 ms for $h_f=10^{-8}$ m when the Hamaker constant $A_H$ is $10^{-20}$ J. This rupture time increases with the film thickness $h$. As aforementioned, when the equilibrium thickness is larger than $10^{-8}$ m due to the nanoparticle-induced disjoining pressures, the rupture time will be elongated and possibly become comparable to the drainage time. In addition to the influences of disjoining pressures, Rio and Biance (2014) proposed that the presence of impurities can also limit the film rupture by the following two mechanisms: damping the fluctuations and providing an energy barrier.

*Fluctuation damping*

![Figure 5.31: Corrugations of bubble interfaces: (a) Without the adsorption of nanoparticles; (b) With the adsorption of nanoparticles (Rio and Biance, 2014).](image)

Bergeron (1997) experimentally investigated the influences of surfactants on the liquid film stability via the Wilhelmy method using a rectangular “open-frame” probe and the porous plate technique. He found the energetic cost associated with thermal fluctuation is increased by the elasticity of the surfactant layer at the gas-liquid interface. This effect tends to decrease the probability of spatial fluctuations. Blute et al. (2007) found silica nanoparticles (5~40 nm) and surfactants have similar effects on increasing the surface elasticity. When the interface is gradually adsorbed by nanoparticles and become rigid, the surface elasticity can exceed the surface tension and reduce the probability of expansion of a fluctuation (Figure 5.31(b)). According to Rio and Biance
(Rio and Biance, 2014), this effect, which is named Gibbs-Maragoni effect, is the common mechanism describing how the presence of impurities can reduce the rupture and increase the stability of the liquid film.

**Energy barrier**

Through a theoretical analysis of the nucleation of a hole in the thin liquid film, Wennerström et al. (1997) found the large curvature energy (a part of surface free energy) of interfaces covered by surfactants also helps stabilizing the thin liquid films. The nucleation of a hole in a thin film is associated with a large curvature, which has an energetic cost that increases the energetic barrier to overcome for rupture (Rio and Biance, 2014). This energy is larger when the surfactants are attached to the interface. Similar explanations could also be found in Timothy et al.’s study (Hunter et al., 2008) where the role of particles in stabilising foams was investigated. It was presented that in the rupture stage an energy barrier must be overcome to form a critical sized hole in the liquid film. Thus the stability of the film can be considered in line with the energy required for the hole formation. Because of the high free energies involved with strongly adsorbed particles, they are far more likely to be laterally moved along the contact interface, rather than expelled into the open liquid. Thus the hole formation and expansion with the existence of nanoparticles can be much more difficult, which consequently elongates the film rupture time.

When the film rupture time in nanoparticle-containing system is sufficiently long and becomes comparable to the film drainage time, it should be incorporated in Equation 5.29 to calculate the coalescence efficiency. However, the real situation is very complicated. As pointed out by Rio and Biance (2014), the film rupture even can be stochastic, if the drainage time is smaller than the time necessary to develop an instability. In view of the fact that all of the above-mentioned mechanisms are closely related to the nanoparticle layer at the bubble interface, more details of the structure of this layer are needed. In recent years, it was found that different orientation and packing structures of nanoparticles can be generated by controlling the nanoparticle aspect ratio, surface properties, concentration and solvent evaporation rates (Böker, He et al., 2007). Moreover, for nonspherical particles, their shape also plays an important
role. For instance, rodlike particles achieve an end-to-end registry of particle faces (Lewandowski et al., 2009), whereas charged ellipsoids can assemble into complex triangular lattices (Madivala et al., 2009). All of these influencing factors may indirectly but profoundly affect the interactions of bubble-liquid and bubble-bubble by creating various structures of nanoparticle layer at bubble interfaces. Further studies are still urgently needed in this area.

5.2.5 Summary

A comprehensive literature review was conducted, which demonstrated that the modification of flow structures is closely related to the changes of bubble-liquid and bubble-bubble interactions induced by the spontaneous nanoparticle adsorption on the bubble surface. The adsorbed nanoparticles make a bubble behave somewhere between a clean bubble and a solid particle. As a result, flow separation occurs and a slanted wake region forms behind the nanoparticle-adsorbed bubble at a small Reynolds number. Both pressure and viscous stress on the bubble interface become asymmetrically distributed due to the nanoparticle surface concentration. In addition, the interactions between nanoparticles such as electrostatic double layer force and steric repulsion force can not only resist the approach of two bubbles, but also hinder the fluctuation of the liquid film. With all of the above changes, the following four results are obtained:

1. The lift force acting on a nanoparticle-coated bubble reverses its direction at a smaller bubble diameter.
2. The drag force increases and enters the distorted regime at a smaller bubble Reynolds number.
3. The thinning process of the liquid film slows down and consequently the film drainage time is elongated.
4. The liquid film is less likely to rupture and the rupture time becomes comparable to the drainage time.

It was, therefore, concluded that the key task when modelling the bubbly flows containing nanoparticles is to formulate the lift force, drag force and film drainage time and rupture time.
Chapter 6

Conclusions

Since nanofluids were proposed and named for the first time by Choi and Eastman (1995), an increasing number of experiments on nanofluids’ properties and performances have been conducted. Compared with those of pure liquids, dilute nanofluids present similar thermo-physical properties but their bubbly flows exhibit dramatically changed bubble characteristics and significantly improved heat transfer performances. However, there remain two major gaps which hinder the further industry application of nanofluids. Because of the inherent complexity, accurate description of the boiling heat transfer and efficient prediction of the heat transfer coefficient (HTC) are still difficult. An in-depth understanding of the heated surface characteristics and bubble hydrodynamics in the near-wall region for both pool and flow boiling of nanofluids is urgently needed. Besides the lack of insight into the heat transfer in nanofluids, the absence of study in two-phase flow structures of their bubbly flows is another gap that needs to be filled, especially for flow boiling. Recently, with the development of computer technology and computation algorithm, Computational Fluid Dynamics (CFD) provides an alternative method to bridge these two gaps.

With the help of CFD, a parametric study of the heat flux partitioning (HFP) model for nucleate boiling of nanofluids was conducted in this study. It was found the surface modifications induced by nanoparticle deposition which were not observed in nucleate boiling of pure liquids is the main cause of the dramatic change of bubble
nucleation characteristics and heat transfer performance. The surface wettability enhancement induced by nanoparticle deposition, among the other parameters had the most significant effect on bubble nucleation on the nanoparticle-deposited heater surface. Therefore, in this thesis new closure correlations were incorporated to characterize the surface modifications and their effects on bubble nucleation and departure when modeling nucleate boiling of nanofluids. A more feasible and mechanistic approach than the classic Rohsenow correlation to predict nucleate boiling of nanofluids was also provided. The HFP model was further improved by containing an additional HFP component that accounts for the heat transfer by the nanoparticle Brownian motion in the microlayer. Due to the continuously increased nanoparticle concentration in the microlayer, heat transfer by the Brownian motion of nanoparticles in the microlayer becomes an important mechanism of heat removal from the heater surfaces boiling in nanofluids. Numerical computations were then conducted using both the new and classic HFP models. The numerical results were analyzed and compared against the experimental data available in the literature. The new HFP model achieved a better agreement with the experimental data than the classic HFP model, especially when the applied heat flux is high. This indicates that the active site density available on the heater surface plays a crucial role in determining the significance of nanoparticle Brownian motion. For dilute nanofluids, the heat transfer due to nanoparticle Brownian motion is positively affected by the bulk concentration and negatively influenced by the nanoparticle size. An increased bulk concentration or a decreased nanoparticle size would enhance the significance of nanoparticle Brownian motion in heat removal. Comparatively, the nanoparticle material does not have much impact on the heat transfer due to the nanoparticle Brownian motion.

In this thesis, the flow structures of bubbly flows of air-water and air-nanofluid were also numerically investigated using the two-fluid model and the MUSIG model, respectively. Comparison of the numerical results against the experimental data available in the literature revealed that the both the above two models agreed well with the experimental data of air-water bubbly flows, but needed substantial improvement in order to achieve an effective modelling of air-nanofluid bubbly flows. The effects of nanoparticles on the interfacial behaviours and interphase transport mechanisms were
analysed based on the experimental observations in the literature. Although the addition of a small amount of nanoparticles into the base liquid does not cause measurable changes in the liquid properties, the spontaneous nanoparticle adsorption at the interface could significantly change the interfacial behaviours of the air bubbles. This was supposed to be the major reason responsible for the distinctly changed two-phase flow characteristics (e.g., smaller bubble size) of air-nanofluid bubbly flows than those of air-water flows. The spontaneous assembly of nanoparticles at the bubble interface significantly changes the interface rigidity and mobility. As a result, the interfacial drag force is increased and the role of lift force with increasing bubble size is modified. It was proven that the positive-to-negative reversal of the lift force occurs at a smaller bubble size in nanofluids compared to that in pure water. The layer of nanoparticles at the bubble surface hinders bubble coalescence by forming a physical barrier and restricting the mobility of the surface. The thinning process of the liquid film trapped between two colliding bubbles slows down, resulting in a longer bubble drainage time. However, the mechanisms responsible for the elongated drainage time are still yet to be uncovered. As the governing equations are still applicable to nanofluids, the most important task when modelling air-nanofluid bubbly flows using the two-fluid model is to formulate the interphase transport terms in order to take into account the specific features induced by the existence of particles.

In the last section of this thesis, the effects of nanoparticle adsorption at bubble interfaces on the two-phase flow behaviours were analysed mechanistically. Due to the adsorption of nanoparticles at bubble interfaces, the slip conditions and properties of the bubble interfaces are significantly changed. The internal circulation is suppressed, leading to an increased shear drag. Moreover, when a bubble is covered with nanoparticles, it would partially behave like a rigid sphere and develop a rotation movement. As a result, flow separation occurs and a slanted wake region forms behind the nanoparticle-adsorbed bubble at a small Reynolds number. This slanted wake would generate a lateral force pointing towards the pipe centre and consequently make the positive-to-negative reversion of the lift force occur at a much smaller bubble diameter. The slanted wake would also make bubbles in the viscous regime experience a drag force similar to that in the distorted regime, which causes the viscous-to-
distorted transition point to occur at a smaller bubble Reynolds number. Meanwhile, both pressure and viscous stress on the bubble interface become asymmetrically distributed due to the nanoparticle surface concentration. In addition, the interactions between nanoparticles such as electrostatic double layer force and steric repulsion force can not only resist the approach of two bubbles, but also hinder the fluctuation of the liquid film. The wake significantly alters the role of lift force and drag force. It is crucial to reformulate the interfacial forces when modelling nanofluid bubbly flows using the two-fluid model. The thinning process of the liquid film slows down and consequently the film drainage time is elongated. The liquid film is less likely to rupture and the rupture time becomes comparable to the drainage time.

However, theoretical modelling of bubbly systems of nanoparticle-liquid mixtures remains very challenging due to the difficulties in formulating the modified bubble behaviours induced by the adsorbed nanoparticles. Traditional two-phase flow theories seem to have encountered a bottleneck. Alternatively, particle-based methods such as molecular dynamics, Brownian dynamics, dissipative particle dynamics and Mente Carlo simulations may be capable of achieving an insight into the embedded physics and generating promising closure models for the two-phase flow models.
Bibliography


