1-1-2008

Experimental measurement of Soret coefficient and numerical study on effect of g-jitter in liquid mixtures

Pouyan Ezzatian
Ryerson University

Follow this and additional works at: <http://digitalcommons.ryerson.ca/dissertations>

Part of the [Mechanical Engineering Commons](#)

Recommended Citation

Ezzatian, Pouyan, "Experimental measurement of Soret coefficient and numerical study on effect of g-jitter in liquid mixtures" (2008). *Theses and dissertations*. Paper 601.

This Thesis is brought to you for free and open access by Digital Commons @ Ryerson. It has been accepted for inclusion in Theses and dissertations by an authorized administrator of Digital Commons @ Ryerson. For more information, please contact bcameron@ryerson.ca.

QC
312
T55
E95
2008

EXPERIMENTAL MEASUREMENT OF SORET COEFFICIENT AND NUMERICAL STUDY ON EFFECT OF G-JITTER IN LIQUID MIXTURES

By

Pouyan Ezzatian
B.Eng. Aerospace Engineering
Ryerson University, 2004

A Thesis

Presented to Ryerson University

In partial fulfillment of the requirement for the degree of

Master of Applied Science

In the Program of

Mechanical Engineering

Toronto, Ontario, Canada, 2008

© Pouyan Ezzatian, 2008

Author's Declaration

I, Pouyan Ezzatian, hereby declare that I am the sole author of this thesis.

I authorize Ryerson University to lend this thesis to other institutions or individuals for the purpose of scholarly research.

I further authorize Ryerson University to reproduce this thesis by photocopying or by other means, in total or in part, at the request of their institutions or individuals for the purpose of scholarly research.

Borrower's Page

Ryerson University requires the signatures of all persons using or photocopying this thesis. Please sign below, and give address and date.

[illegible]

Abstract

Experimental measurement of Soret coefficient and effect of g-jitter in liquid mixtures

Master of Applied Science – Mechanical Engineering, 2009

Pouyan Ezzatian

School of Graduate Studies, Ryerson University

A ground based experiment using digital optical interferometry was conducted to measure the Soret and thermodiffusion coefficients of a liquid binary mixture of water-isopropanol with different mass fractions in a rectangular glass cell. The cell was subjected to a temperature gradient of 6°C , and the entire experimental setup was placed inside a heat insulation box. The results showed good agreement with published results and established that the method of measurement is a competitive and accurate method to measure the transport coefficients in binary liquid mixtures. In preparation for future IVIDIL (influence of vibrations on diffusion in liquids) experiments on board the International Space Station, a numerical analysis for the mixture of water-10% isopropanol was performed to determine the effects of g-jitter on the flow inside a cylindrical model subject to a temperature gradient of 10°C . The data for g-jitter, Soret and thermodiffusion coefficients were obtained from FOTON-12 and used in the analysis. A comparison between zero-gravity, micro-gravity and g-jitter scenarios showed that the presence of g-jitter does affect the flow inside the cell. Temperature and concentration times to reach the steady state were increased.

Acknowledgements

I would like to express my gratitude to Professor Ziad Saghir – head of the Microgravity Laboratory in the Mechanical Department, Ryerson University, Toronto, Canada – for his support, guidance and encouragement during the course of my research and the completion of this report.

I would also like to thank Professor Valentina Shevtsova and Dr. Alexander Mialdun – at the Microgravity Research Centre in the Faculty of Engineering, Université Libre Bruxelles, Brussels, Belgium – for their guidance and support during my three month visit to conduct experiments.

In addition, I would like to acknowledge the help and useful suggestions of my colleagues Dr. Yu (Claire) Yan, Mehdi M. Shemirani (PhD Candidate) and Tawfiq Jaber (PhD Candidate).

Dedication

I would like to dedicate this thesis to my parents Mohammad and Mansoureh, and my brother Payam as a token of appreciation for their patience, support, love and encouragement throughout the completion of this thesis.

Table of Contents

AUTHOR’S DECLARATION	II
BORROWER’S PAGE	III
ABSTRACT	IV
ACKNOWLEDGEMENTS	V
DEDICATION	VI
TABLE OF CONTENTS	VII
LIST OF TABLES	X
LIST OF FIGURES	XI
NOMENCLATURE	XIV
CHAPTER 1. INTRODUCTION AND LITERATURE REVIEW	1
1.1 NUMERICAL SEGMENT.....	1
1.1.1 Introduction.....	1
1.1.2 Literature Review	2
1.2 EXPERIMENTAL SEGMENT.....	9
1.2.1 Introduction.....	9
1.2.2 Literature Review	10
1.2.3 Applications in Industry.....	22
1.3 RESEARCH OBJECTIVE	23
1.4 THESIS ORGANIZATION	24
CHAPTER 2. NUMERICAL AND EXPERIMENTAL FORMULATION	25
2.1 INTRODUCTION	25
2.2 GOVERNING EQUATIONS USED WITH THE NUMERICAL PART	25
2.2.1 Navier-Stokes Equations	25
2.2.2 Energy Equation	27
2.2.3 Species Conservation Equation	27
2.3 BOUNDARY AND INITIAL CONDITIONS	28
2.4 SOLUTION TECHNIQUE AND CONVERGENCE CRITERIA.....	30
2.5 COMSOL MULTIPHYSICS	33
2.6 GOVERNING EQUATIONS FOR THE EXPERIMENTAL PART	33
2.6.1 Phase change	34
2.6.2 Change of refractive index	34
2.6.3 Fitting curve equations.....	34
2.6.4 Two-dimensional Fast Fourier Transform	35
CHAPTER 3. EXPERIMENTAL MEASUREMENT OF SORET COEFFICIENTS FOR LIQUID MIXTURES	36
3.1 EXPERIMENTAL SETUP AND EQUIPMENT PROPERTIES.....	36
3.2 EXPERIMENTAL PROCEDURE	40
3.2.1. Experimental preparation	40

3.2.2. Processing of interferogram.....	43
3.3 RESULTS AND DISCUSSION	49
3.3.1. Method validation against Fontainebleau benchmark	49
3.3.1.1 IBB- $C_{12}H_{26}$ Mixture	50
3.3.1.2 IBB-THN Mixture	53
3.3.1.3 THN- $C_{12}H_{26}$ (Dodecane) Mixture	55
3.3.1.4 Dodecane-THN Mixture.....	57
3.3.2. Additional validation with ethanol-water mixtures	59
3.3.3. Defining method limitations.....	63
3.3.4. Isopropanol-water mixtures.....	64
3.3.4.1 Isopropanol – 50wt% water Mixture	64
3.3.4.2 Isopropanol-1 wt% water	67
3.3.4.3 Isopropanol-5 wt% water Mixture	68
3.3.4.4 Isopropanol-15 wt% water Mixture	69
3.3.4.5 Isopropanol-80 wt% water Mixture	70
3.3.4.6 Isopropanol-70 wt% water Mixture	71
3.4 SUMMARY.....	73
CHAPTER 4. STUDY OF THE EFFECTS OF G-JITTER IN LIQUIDS IN THE PRESENCE OF THERMODIFFUSION	75
4.1 INTRODUCTION	75
4.2 MODEL DESCRIPTION	75
4.3 MESH ANALYSIS	77
4.4 NUMERICAL RESULTS	80
4.4.1 Zero gravity case	80
4.4.1.1 Temperature analysis	80
4.4.1.2 Concentration analysis	84
4.4.2 Two-dimensional Micro-gravity case	87
4.4.2.1 Velocity contour	89
4.4.3 Three-dimensional G-jitter case	90
4.4.3.1 Temperature analysis	90
4.4.3.2 Concentration analysis	92
4.4.3.3 Velocity analysis	95
4.5 SUMMARY.....	98
CHAPTER 5. CONCLUSION AND FUTURE WORK	99
5.1 SUMMARY.....	99
5.3 FUTURE WORK	100
REFERENCES	101
APPENDICES	105
APPENDIX A: COMSOL 3D G-JITTER MODEL INPUT FILE	105
A.1. TABLE OF CONTENTS	105
A.2. MODEL PROPERTIES	105
A.2.1. MODEL DESCRIPTION	106
A.3. CONSTANTS.....	106
A.4. GEOMETRY	108

A.4.1. GEOM2.....	109
A.4.1.1. Point mode.....	109
A.4.1.2. Edge mode.....	109
A.4.1.3. Boundary mode.....	110
A.4.1.4. Subdomain mode.....	110
A.5. GEOM2.....	111
A.5.1. EXPRESSIONS.....	111
A.5.1.1. Subdomain Expressions.....	111
A.5.2. MESH.....	112
A.5.2.1. Mesh Statistics.....	112
A.5.3. APPLICATION MODE: CONVECTION AND CONDUCTION (CC).....	113
A.5.3.1. Application Mode Properties.....	113
A.5.3.2. Variables.....	113
A.5.3.3. Boundary Settings.....	114
A.5.3.4. Subdomain Settings.....	114
A.5.4. APPLICATION MODE: INCOMPRESSIBLE NAVIER-STOKES (NS).....	114
A.5.4.1. Application Mode Properties.....	115
A.5.4.2. Variables.....	115
A.5.4.3. Point Settings.....	115
A.5.4.4. Boundary Settings.....	115
A.5.4.5. Subdomain Settings.....	115
A.5.5. APPLICATION MODE: PDE, GENERAL FORM (G).....	116
A.5.5.1. Application Mode Properties.....	116
A.5.5.2. Variables.....	116
A.5.5.3. Boundary Settings.....	117
A.5.5.4. Subdomain Settings.....	117
A.6. SOLVER SETTINGS.....	117
A.6.1. DIRECT (UMFPACK).....	118
A.6.2. TIME STEPPING.....	118
A.6.3. ADVANCED.....	119
A.7. POSTPROCESSING.....	120
APPENDIX B: CELL TECHNICAL DRAWING (EXPERIMENTAL).....	121
APPENDIX C: CHEMICAL PROPERTIES.....	122
1,2,3,4-Tetrahydronaphthalene.....	122
Isobutylbenzene.....	123
Dodecane.....	124
APPENDIX D: MATLAB CODE.....	127
APPENDIX E: CELL TECHNICAL DRAWING (NUMERICAL).....	129

List of Tables

Table 2.1: Thermo-physical properties of water-isopropanol (90-10 weight %)	30
Table 2.2: Fourier coefficients for Foton-TRAMP [9]	32
Table 3.1: Individual results for Fontainebleau binary mixtures ($T_0=25^{\circ}\text{C}$, $c_0=0.5$), Platten et al. [32]	50
Table 3.2: Proposed benchmark values for Fontainebleau mixtures, Platten et al. [32]	50
Table 4.1: Mesh statistics of two-dimensional model	77
Table 4.2: Three-dimensional mesh statistics	78

List of Figures

Figure 2.1: Three-dimensional model (edge mode).....	28
Figure 3.1: Schematic of experimental setup	36
Figure 3.2: Optical experimental setup based on Mach-Zehnder interferometry.....	37
Figure 3.3: SolidWorks drawing of cell, with copper plates installed	38
Figure 3.4: Picture of the actual experimental cell and hardware	39
Figure 3.5: Experimental setup showing cell filling arrangement and apparatus.....	42
Figure 3.6: Typical (cropped) raw interferogram obtained during experiment data acquisition.....	43
Figure 3.7: Principle of the image processing procedure using the two-dimensional FFT	44
Figure 3.8: Phase difference plots in the X and Z directions vs. time. The circled data point is selected as the reference image	46
Figure 3.9: Concentration distribution of IBB-THN. Small Perturbations visible in all four corners	47
Figure 3.10: Plots of Standard concentration deviation, diffusion, Soret and initial concentration change vs. layer thickness, respectively.....	48
Figure 3.11: Thermodiffusion plot of concentration difference vs time of IBB-Dodecane	51
Figure 3.12: Diffusion plot of concentration difference vs time of IBB-Dodecane	51
Figure 3.13: Comparison of present experimental diffusion values with other literature results for Dodecane-IBB	52
Figure 3.14: Comparison of present experimental Soret values with other literature results for Dodecane-IBB	52
Figure 3.15: Thermodiffusion plot of concentration difference vs. time of IBB-THN.....	53
Figure 3.16: Diffusion plot of concentration difference vs time of IBB-THN.....	54
Figure 3.17: Comparison of present experimental diffusion values with other literature results for IBB-THN	54
Figure 3.18: Comparison of present experimental Soret values with other literature results for IBB-THN	55
Figure 3.19: Thermodiffusion plot of concentration difference vs time of THN-Dodecane.....	55
Figure 3.20: Diffusion plot of concentration difference vs time of THN-Dodecane	56
Figure 3.21: Comparison of present experimental diffusion values with other literature results for Dodecane-THN	56

Figure 3.22: Comparison of present experimental Soret values with other literature results for Dodecane-THN.....	57
Figure 3.23: Concentration change vs. thermodiffusion time for dodecane-40 wt% THN.....	58
Figure 3.24: Concentration change vs. diffusion relaxation time for dodecane-40 wt% THN	59
Figure 3.25: Concentration change vs time of water 50 wt%-ethanol	60
Figure 3.26: Diffusion plot of concentration difference vs time of Ethanol-50wt% water	61
Figure 3.27: Plot of various diffusion coefficients vs. different water-ethanol mass fractions.....	62
Figure 3.28: Plot of various Soret coefficients vs. different water-ethanol mass fractions.....	63
Figure 3.29: Water 90 wt%-ethanol concentration plot	64
Figure 3.30: Thermodiffusion plot of concentration change vs. time for water-isopropanol mixture	65
Figure 3.31: Soret coefficient of water-isopropanol mixture vs. water mass fraction showing present result.....	66
Figure 3.32: Diffusion coefficient of water-isopropanol mixture as a function of water mass fraction showing present result.....	67
Figure 3.33: Thermodiffusion plot of concentration change vs. time for water-isopropanol mixture	68
Figure 3.34: Thermodiffusion plot of concentration change vs. time for isopropanol-5wt% water mixture	69
Figure 3.35: Thermodiffusion plot of concentration change vs. time for isopropanol-15wt% water mixture.....	70
Figure 3.36: Thermodiffusion plot of concentration change vs. time for isopropanol-70wt% water mixture.....	71
Figure 3.37: Present results of Soret coefficient of water-isopropanol mixture vs. water mass fraction compared to literature results	72
Figure 3.38: Present results of diffusion coefficient of water-isopropanol mixture vs. water mass fraction compared to literature results	72
Figure 4.1: Numerical model generated in COMSOL	76
Figure 4.2: Three-dimensional model.....	77
Figure 4.3: Three-dimensional model.....	79
Figure 4.4: Three-dimensional Mesh quality plot.....	80
Figure 4.5: Two-dimensional Temperature and concentration distribution at time 300 hours	81
Figure 4.6a-c: Temperature vs time at points 1:(0.0135,0.006), 2:(0.02,0.006) & 3:(0.0265,0.006) respectively	82
Figure 4.7: Two-dimensional model showing locations of 3 points	83
Figure 4.8: Temperature across the centre line of the model (x in metres) at four different times	84

57	Figure 4.9: Two-dimensional model showing 4 point where concentration data was taken.....
58	Figure 4.10: Two-dimensional 0-g concentration differences across the model, and average
59	concentration difference
60	Figure 4.11: Two-dimensional microgravity model temperature and concentration distribution at 200 hours
61	Figure 4.12: Temperature versus time at point $(x,y) = (0.0265, 0.006)$
62	Figure 4.13: Temperature across the centre line of the model (x in metres) at different times.....
63	Figure 4.14: Velocity contour plot in x-direction for two-dimensional model
64	Figure 4.15a-c: Temperature vs time for three different points along the centre x-axis
65	Figure 4.16: Temperature distribution of three-dimensional g-jitter model at 300 hours
66	Figure 4.17a-b: Three-dimensional g-jitter concentration differences across the model, and average concentrati
ent	difference
67	Figure 4.18: Comparison plot of the difference of concentration averages between the hot and cold sides, for th
68	two-dimensional 0-g and three-dimensional g-jitter cases
69	Figure 4.19a: X-velocity vs time for three-dimensional g-jitter from time 291.6-300 hours
70	Figure 4.19b: Zoomed-in view of X-velocity vs time for three-dimensional g-jitter from time 291.6-300 hours..
71	Figure 4.20a: Y-velocity vs time for three-dimensional g-jitter from time 291.6-300 hours
72	Figure 4.20b: Zoomed-in view of Y-velocity vs time for three-dimensional g-jitter from time 291.6-300 hours..
72	Figure 4.21a: Z-velocity vs time for three-dimensional g-jitter from time 291.6-300 hours.....
72	Figure 4.21b: Zoomed-in view of Z-velocity vs time for three-dimensional g-jitter from time 291.6-300 hours..
76	Figure B.1: Technical drawing of the experimental glass cell.....
77	Figure E.1: Technical drawing of the cell used in numerical analysis

Nomenclature

a_0, a_j	Fourier coefficients [m/s^2]
b_j	Fourier coefficient [m/s^2]
C, C_0	Concentration (mass) and initial concentration at time zero respectively
C_p	Specific heat [$\text{J/Kg } ^\circ\text{K}$]
D_T	Thermodiffusion coefficient [m^2/s]
D_C	Molecular diffusion coefficient [m^2/s]
f_j	Frequency in the Fourier series, for $j = 1, \dots, m$ [Hz]
H	Height [m]
$F(u,v)$	Fourier transform
g	Gravity [m^2/s]
$J_{x,y,z}$	Diffusion flux in the X, Y and Z-directions
K	Degree Kelvin
L	Length [m], or thickness of liquid [m]
n	image number
p, p_0	Pressure and initial pressure at time zero, respectively [Pa]
S_T	Thermal Soret coefficient [$1/^\circ\text{K}$]
t	time [s]
T, T_0	Temperature and initial temperature at time zero, respectively [$^\circ\text{K}$]
u	Velocity in the X-direction [m/s]
v	Velocity in the Y-direction [m/s]
w	Velocity in the Z-direction [m/s]
W	Width [m]

Greek symbols:

α	Thermal diffusivity [m^2/s]
β_c	Volumetric concentration coefficient
β_T	Thermal expansion coefficient [$1/^\circ\text{K}$]
κ	Thermal conductivity [W/m K]
λ	Wavelength [m]
μ	Dynamic viscosity [Kg/m s]
ν	kinematic viscosity [m^2/s]
ρ, ρ_0	Density and initial density at time zero, respectively [Kg/m s]
τ_r	Relaxation time [s]

Subscripts:

0	Reference state or initial state
cold	At the cold wall
hot	At the hot wall
x	X-direction in the Cartesian coordinate system
y	Y-direction in the Cartesian coordinate system
z	Z-direction in the Cartesian coordinate system

Abbreviations:

2D	Two-dimensional
3D	Three-dimensional
FFT	Fast Fourier Transform
IBB	Isobutylbenzene
ISS	International Space Station
IVIDIL	Influence of vibrations on diffusion in liquids
THN	1,2,3,4-Tetrahydronaphthalene

Chapter 1. Introduction and Literature Review

1.1 Numerical Segment

1.1.1 Introduction

The net movement of particles moving from an area of high concentration to an area of lower concentration is called 'diffusion'. Therefore, the net movement of molecules moving from an area of high concentration to an area of lower concentration is called molecular diffusion. There will be some movement from the lower area of concentration to the higher area of concentration as well; however, this is at a much lower rate. This net movement in turns creates a concentration gradient in the mixture, which over time decreases as the concentration in both areas becomes equal.

The formation of a mass flux in the presence of a temperature gradient in a binary mixture is referred to as the Soret effect or thermal diffusion. This separation phenomenon may occur for both liquid and gaseous mixtures. Although the Soret effect is considered to be small, it is an important component in the analysis of hydrocarbons in oil reservoirs.

The Soret effect is measured using the Soret coefficient, defined as a ratio of the thermal and molecular diffusion coefficients. The Soret coefficient essentially measures the separation of the components in a mixture which is subjected to a temperature gradient. There are several different experimental approaches to determine the Soret coefficient (such as the thermo-gravitational method and the microgravity method). Hang and Li [1] determined through experimental results that as the viscosity of a mixture increases, so does the Soret coefficient. This result therefore implies that there is a relation between the Soret coefficient and molecular interactions in

mixtures. This in turn affects the sign change of the Soret coefficient as well, depending on the molar fraction of a component.

Given the importance of the Soret effect, efforts have been made to effectively develop both experimental and theoretical models. However, due to buoyancy induced convection on Earth, these efforts have been greatly hindered, and thus the measurement is very difficult. The ideal scenario would be a zero-gravity environment, yet this is also challenging. There have been experiments conducted in micro-gravity environments, such as the International Space Station (ISS) and the Russian satellite Foton-12. Even in these environments, however, there are still residual gravitational forces and instabilities. These static and oscillatory residual accelerations are termed 'g-jitter' and are caused, for example, by the activation of thrusters, crew movement, or on board machinery, etc. Although g-jitter forces are much weaker than those on Earth, they still have a range of frequency and amplitude, and therefore their effect on convection must be studied in order to obtain accurate results.

1.1.2 Literature Review

Previous studies regarding the effects of g-jitter on thermal diffusion have been published by several researchers. Haddon and Riley [2], Davidson [3] and Merkin [4] studied the effect of acceleration fluctuations in a liquid. Their findings established that the g-jitter induced convection resulted in significantly higher heat transport, when compared to the diffusive state. Chacha et al. [5] performed a 2D numerical analysis for a binary mixture, studying the effect of density variation and vibrational instability in a microgravity environment. The model was a rigid rectangular cavity filled with methane and butane, subjected to g-jitter. The g-jitter acceleration was studied in two orientations – perpendicular and parallel to the thermal gradient. In terms of boundary conditions, the top and bottom walls were allowed heat exchange with the environment.

environment. The vertical walls were set to a hot and cold temperature, thus creating a thermal gradient. The principles of mass, momentum and energy conservation were applied to the model. The numerical approach used the appropriate boundary conditions to solve for the conservation equations, using the Galerkin finite difference method for a variable density. Three different cases were investigated: zero gravity, g-jitter in the x-direction, and g-jitter in the y-direction. The results presented were at the time when the thermal steady state was reached. Their results showed that the temperature profile distortions increased when a parallel g-jitter was added to the residual gravity. They also proved that depending on the orientation of the vibration and residual gravity vector relative to the density gradient direction, both residual gravity and g-jitter may be beneficial or detrimental to achieving purely diffusive conditions.

Savino [6] performed a numerical study for two different experiments that show sensitivity to residual accelerations: one was the measurement of thermal conductivity in high Prandtl numbers, the other was the measurement of the thermodiffusion coefficient in binary mixtures. The residual gravity data used were those obtained experimentally on board the ISS. The second experiment, involved the measurement of the Soret coefficient in a metal alloy, subjected to a temperature gradient across a cell. The analysis showed that orienting the residual-g parallel with the density gradient resulted in a reduction of the conductive instabilities.

Yan et al. [7] carried out a 3D numerical study on the effects of low frequency g-jitter on thermodiffusion. The mixture was water-isopropanol (90:10 weight percentages) in a cubic cell subjected to lateral heating and several vibration conditions, with low frequencies ranging from 0.001 Hz to 0.025 Hz. The cubical cavity consisted of two opposite side walls at different constant temperatures, one being the hot wall, the other the cold wall. All other walls of the model were thermally insulated. The applied static and oscillatory gravities were applied in the

vertical (Z) direction. The governing equations for this double diffusion convection process were the mass, momentum, energy and species conservation equations. The Boussinesq approximation was applied to solve the governing equations. In terms of the boundary conditions, the no-slip condition was applied to the side walls; in addition, there were no chemical reactions and no mass flux inside or through the cavity. The model was solved using the finite volume method, with the iteration process starting by using guessed initial velocity and pressure fields. Using the above method and solving the continuity equation, a pressure correction field is obtained and subsequently the velocity and pressure fields are updated. The iteration then continued until convergence is reached. Different cases were investigated to study their effects on the diffusion process: static residual gravity, pure oscillatory gravity and a combination of the aforementioned. The results yielded several conclusions. For one, it was determined that the combined effects of vibrations on diffusion were a combination of effects caused by each individual g-jitter component. Adding complication to this finding is that the interactions between the static and oscillatory components are non-linear. In the case of the static residual gravity, it was determined that, depending on the magnitude of the residual gravity, static residual gravity could cause a strong fluid flow, resulting in an unstable Soret separation, thus affecting the accuracy of the diffusion measurement. Lastly, in the case of the oscillatory gravity, the study found that the effect of convection at low frequency g-jitter greatly influenced (negatively) the concentration and temperature distributions when compared to high frequency g-jitter.

Furthermore, Yan et al. [8] conducted a comparative study of the thermodiffusion process in a ternary mixture containing methane, dodecane and n-butane. A 3D numerical model was developed and data from experiments aboard the ISS were compared to theoretical results to determine the influence of microgravity on thermodiffusion. Six scenarios were investigated, one being

ideal zero-gravity scenario, and the remaining five scenarios using acceleration data measured onboard the ISS. The scenarios involved “best” and “worst” conditions, for when the acceleration values were at a minimum and maximum, respectively, related to the active or inactive status of the crew. The model was subjected to different levels, frequencies and alignments of g-jitter. The model used was a rectangular cavity, with opposite heated side walls, one side hot and the other cold, thus creating a temperature gradient. The full transient Navier Stokes, momentum and energy equations were solved using a finite volume method. The boundary conditions applied were the no-slip and no-flow condition at the walls, and all walls (except for the heated walls) were thermally insulated. In addition, no chemical reactions took place inside the cavity, and no mass flux existed through the walls. The above boundary conditions were applied and the system of equations was solved for in the same manner discussed in the previous mentioned review of Yan et al. [7]. For all scenarios studied, the temperature profile shows small deviations from the zero-gravity linear distribution case. It was determined, however, that the differences were negligible due to small g-jitter magnitudes. The lateral temperature gradient did result in a component separation, with dodecane and n-butane migrated to the cold wall, and methane at the hot wall. This separation caused a non-uniform mixture. The results concluded that the presence of g-jitters negatively affected the diffusion process. However, the effects were low in magnitude, and therefore the negative impact was relatively acceptable. It was also concluded that the ISS is a suitable platform for microgravity experiments. In addition, in order to control the magnitude of g-jitters, it was recommended to introduce a threshold value of $10\mu\text{g}$, above which the experimental results could be negatively affected.

Yan and Saghir [9] performed a similar study using a ternary mixture of n-butane, methane and dodecane in a rectangular cavity, undergoing lateral heating, and various g-jitter levels. These

results were obtained from experimental data aboard FOTON-12 and the ISS and compared with the ideal zero-gravity condition. The governing equations were the continuity, species, momentum and energy equations, and were solved using the finite volume method. Boundary conditions included the no-slip and no-flow conditions, rigid walls, and all walls were assumed to be rigid except. In addition, all walls except the hot and cold walls were assumed to be thermally insulated. Local properties such as concentration, temperature and diffusion coefficients and the effect of microgravity on these properties were examined. At zero gravity, the temperature was found to vary linearly from the hot side to the cold side, after steady state was reached. In terms of concentration and component distribution, at zero gravity, the separation process was found to be smooth; however, when microgravity was introduced, the separation process was disturbed due to the convective flow. A non-uniform distribution was also observed for molar and thermal diffusion. This was found to be due to the non-uniformity in both concentration and temperature in the cavity. The hot and cold walls showed a high sensitivity to g-jitter, and the centre of the cavity showed the least sensitivity, and hence fluctuations were observed at this location. The authors also studied the effects of location on the results. A comparison was made between the results of the zero gravity case for ISS and FOTON locations. Although deviations in results were observed, and FOTON was determined to be a favoured location, the deviations were so small such that it was concluded both platforms offered a suitable ground for microgravity experiments. In the end, it was noted that the location of the experiment on aboard both platforms is of great importance, in order to keep the effect of g-jitter at a minimum.

Chacha and Saghir [10] studied the effects of g-jitter on the solutal thermodiffusion of a two-dimensional rectangular cavity filled with a binary mixture (methane and n-butane) subjected to a temperature difference on its lateral walls and varying vibration in microgravity environment.

For boundary conditions, the g-jitter was assumed to be perpendicular to the temperature gradient. The vertical walls were set to constant temperatures, one being the hot wall, the other the cold wall. The horizontal walls were allowed to exchange heat with the surrounding environment. All walls were assumed to be rigid and impermeable to matter, and the no-flow and no-slip conditions were applied to all four walls of the cavity. The governing systems of equations were the mass, momentum, energy equations. All physical properties were varying in both time and space, except for the fluid specific heat and thermal conductivity at constant pressure. Like the previous mentioned models, this model was a non-linear, coupled problem. The numerical procedure used was based on the SIMPLE algorithm. Different levels of accelerations were examined: earth gravity condition, residual gravity and microgravity. For the earth gravity scenario, the results yielded a uniform fluid mixture distribution, which was generated by the buoyancy convection. The zero gravity scenario was then introduced which resulted in a symmetrical distribution of butane with respect to the horizontal centre line, and it was also found that butane would migrate to the cold wall in the absence of gravity. The introduction of residual gravity initiated mixing in the cavity. Although the level of residual gravity was very low, it was noted that the buoyancy was affecting the mass diffusion and it was observed that less butane was migrating to the cold wall. A comparison was made between the zero gravity and microgravity scenarios for the butane distribution. It was observed that for the zero gravity case, the distribution was concave and symmetrical. As residual gravity levels were increased, buoyancy forces were observed getting stronger resulting in a disturbed profile. Finally, the effects of time varying amplitude and frequency were examined. They concluded that due to variation in frequency and amplitude (as gravity became time dependent), the mass diffusion was affected by the vibrations. In addition, they determined that the Soret number

oscillates with time when the frequency is the same as the original g-jitter excitation. However, the variation is disturbed by a backflow, and hence its shape is non-sinusoidal.

Chacha et al. [11] also investigated the effects of residual and very low frequency g-jitter on mass diffusion in a ternary mixture. The data for this investigation was obtained from experiments aboard FOTON. The system examined was a three dimensional rectangular cavity filled with a mixture of butane, dodecane and methane. The vertical walls were set to constant temperatures, with one hot wall and the other a cold wall. All walls were considered to be rigid and impermeable to matter. The no-slip, no-flow boundary conditions were also applied. The energy, mass and momentum equations were solved numerically using the control volume method. Once again, all properties vary with time and space except the fluid specific heat and thermal conductivity at constant pressure. As with previous studies, it was noticed that under microgravity conditions, dodecane migrated toward the cold wall. The same behaviour was observed for butane. Methane was observed to migrate toward the hot wall. In addition, it was determined that for a favourable alignment of the mixture with respect to the gravity vector, the onboard vibrations were negligible. In an unfavourable alignment with the residual acceleration, mass differentiation was found to be approximately 20% less when compared to the zero-gravity case. Overall, the authors concluded once again that FOTON was a suitable platform for microgravity experiments.

Shevtsova et al. [12] studied the vibrational effects on thermodiffusion in a binary mixture of water and isopropanol in a cubic cell, undergoing a temperature difference between opposite walls. Vibrations with two different frequencies were investigated and analyzed for component separation due to the Soret effect. The direction of vibrations was perpendicular to the temperature gradient and parallel to the gravity vector. All other walls of the cell were

considered thermally insulated. The no-slip condition was assumed, in addition to all walls being impermeable. For the case of steady residual accelerations with no vibrations, the results showed that the fluid mixture was driven by the buoyancy forces, when the gravity value was set to $10^{-2}g$. In the case of pure oscillatory acceleration, the results showed that temperature, concentration and velocity oscillated locally with the external frequency near the hot wall. However, these oscillations were so small, they could not be measured. Therefore, the Soret separation was determined to be not sensitive to vibrations. Two other cases were examined: one was a combination of the previous scenarios with steady and oscillatory components, and the other was the low frequency vibrations case. They determined that at high and low frequencies, the impact of vibration on flow patterns is different. The heat and mass transport increased when frequency decreased. Temperature and concentration were affected much more when the vibrations were at low frequency. In the event of combined high frequency g-jitter and static gravity, buoyancy was the driving force for the Soret separation. Shevtsova et al. [13] also found that oscillations in the normal direction to the density gradient greatly increased the mass and heat transfer when the oscillatory-g was larger than the stationary-g values.

1.2 Experimental Segment

1.2.1 Introduction

An interesting method used to measure the Soret and the diffusion coefficients in liquids is digital interferometry. Digital interferometry is not a new technique and has been in existence for some years. However, it is gaining popularity. It is widely used for surface deformation and displacement measurements, as well as measurement of the refractivity of objects. By measuring the change in the refractive index, the change in other quantities such as temperature and density can be determined. Typically, interferograms consist of bright and dark fringes. Fringes or fringe

patterns are essentially the phase difference between two overlapping and coherent light which are captured on images [14].

To get a clear picture of the fringes, however, it is necessary to use a high resolution camera for best clarity. Therefore, holograms are recorded by using a Charge Coupled Device (CCD) camera which allow for an interface with the computer. More importantly, they are equipped with a very sensitive thin recording material with high responsivity and a wide spectrum from infrared to ultraviolet [15]. The CCD camera is equipped with photo detectors that sample the signal (two dimensional signals in this case), converts the signal into a digital format and stores it on a frame grabber for further analysis [16]. The detailed physics of operation of a CCD camera will not be further explained here.

In order to extract data from these images, mathematical techniques and analysis must be applied to translate the image into something more sensible. Several types of decoding of optical interferometry exist, such as the two dimensional Fast Fourier Transform (FFT) method. Essentially the method is a phase shift technique, where two images are superimposed to determine the difference. The first image is considered the initial stage, and the second the final stage. This method will be discussed in more detail in the experimental section.

1.2.2 Literature Review

Different experimental setups are used in measurements of the Soret and diffusion coefficients using interferometry.

Li *et al.* [17] developed an interferometric technique to measure - within 0.5% accuracy - the temperature derivative and the refractive index of liquids. The mixtures used in their experiments were mixtures of liquid n-hexane and toluene. The concentration dependence of the refractive

index of the mixtures was measured for n-hexane and toluene and for three mixtures of the binary system for a temperature range of 15 to 45°C. The interferometric setup consisted of a laser beam being transmitted through a converging lens and passing through the centre of the cell containing the mixture. The reflected beams, which created a concentric fringe pattern, were projected onto a screen one meter away, with a 2mm hole in the centre, to allow the central fringe to pass through to a photo diode. For different mass fractions of the mixture and the range of temperatures mentioned, it was evident that the temperature derivative of the refractive index varied with temperature. This result could not be compared to data from other literature because they did not have sufficient accuracy to resolve the temperature dependence of the temperature derivative of the refractive index. The dependence of the concentration on the refractive index was also investigated for toluene and n-hexane. The results obtained for a temperature of 25°C showed good agreement with values obtained in other literature. In fact, the values of temperature derivatives of the refractive index had a higher accuracy than those obtained by other investigators. The temperature derivatives were also compared with theoretical calculations for both liquids. Two equations were investigated: the Looyenga formula and the Lorentz-Lorenz formula. When compared with the experimental results, there was excellent agreement with the Looyenga formula, while there was a 5-10 % deviation with the other formula.

Spagnolo *et al.* [18] developed a Fourier transform algorithm to evaluate interferograms obtained by a TV-speckel interferometer. The method allowed for a point wise calculation of the interference phase. The phase distribution was then combined with computer data with respect to the geometric values of the experimental setup to determine the index of refraction gradients. The speckle patterns produced were recorded by a CCD camera. By rotating the object beam of the interferometer by a certain angle, linear fringes were introduced. The remainder of the experimental setup details was described in another work of the author. As before, the obtained

fringes were subtracted from another set of fringes considered to be at an initial steady state. The developed algorithm was tested both numerically and experimentally. The experimental solvent was LiBr at 25°C. A comparison of the results showed excellent agreement with the theoretical value. The average experimental value was 1.401×10^5 compared to a theoretical value of 1.404×10^5 cm²/s. Thus, it was concluded that the developed algorithm in combination with the interferometric method produced favourable results for the liquid tested at room temperature.

Rashidnia *et al.* [19] developed a common-path interferometer (CPI) system to measure the diffusivity of silicone oils with various viscosities. The interferometer setup consists of a polarized laser beam and a Wollaston prism in combination with an analyzer. The authors compared this method over other interferometer methods (such as point-diffraction interferometry and Schlieren photographic technique) because the CPI has the capability to take quantitative measurements in liquids with high variations in index of refraction. In brief, in this technique, polarized light from a laser passes through the cell and is focused on a Wollaston prism, which then splits the light into two beams that have a slight displacement from each other. A shear element allows the two rays to coincide once they have passed through the element. The two beams interfere with each other, causing a change in the index of refraction and optical path length, which produce interference fringes. The index of refraction gradient is measured when the solvent and liquid is introduced in the cell causing diffusion and changing the shape of the fringes. The method developed for this experiment was used to determine in real time the concentration gradient in the mixing zone. The cell used was made of quartz glass with dimensions 10 x 10 x 45 mm in the x, y and z-directions respectively. The interferograms were captured by a 640 x 480 pixel CCD camera. Diffusivity of several different silicone oil liquid pairs was measured and compared with results from other literature using Wiener's method (where it is required to know the values of the refractive indices of the fluids). This method was found to be the least accurate.

A modified version of the Wiener method and interferometry method measuring the concentration gradient provided good results, with diffusion values such as 5.11 and 5.41×10^{-8} . The exception was for one result for which the diffusion across the mixing interface was found to vary by as much as 36%. It was determined that the assumption of the simple model of diffusion does not hold for this combination of liquid. It was concluded that the CPI method used in this experiment could be used for concentration gradient measurements near moving interfaces for transparent fluids.

Kolodner *et al.* [20] used a laser beam deflection method to measure the Soret coefficient of water-ethanol mixtures, with ethanol weight fractions from 0.02 to 0.39, with temperatures ranging from 10-40°C. The rectangular experimental cell containing the binary mixture is situated between two copper plates, which were subjected to a temperature gradient. The deflected laser beam is sent through the centre of the cell and is received by a detector. The results obtained for the Soret coefficients were satisfactory; however, a comparison with other experimental results was not very informative. Results of the mass diffusion coefficient were plotted as functions of temperature at different concentrations. At a concentration of 0.2587, a high temperature rise was observed. Outside this peak region, the results for the mass diffusion coefficient showed excellent agreement with other experimental literature, for measurements made at 25°C. The authors were not able to provide an explanation for the peak observed.

Zhang *et al.* [21] measured the Soret coefficient of a toluene and n-hexane mixture by determining the horizontal deflection of the laser through the mixture. This is the principle of the beam deflection technique. The experimental cell was different than other experiments in that it contained a longer path length, 6 cm as opposed to 2 to 2.4 cm used by other investigators. The cell, situated between copper plates, was subjected to a vertical temperature gradient and

measurements were taken for a temperature range of 5-45°C, over the entire range of concentrations. The temperatures of the copper plates were controlled by using two water baths. The deflected beam is received by a beam-position detector. To prevent the effect of air convection, the entire experiment was enclosed in a Plexiglas covering. The cells were initially filled with one-component liquids, and the refractive index gradients were measured for each liquid. The overall accuracy was found to be 0.5%, when compared with interferometric measurements taken by Li *et al.* [17]. Therefore, it was concluded that the values for the refractive index gradient were in excellent agreement. The Soret coefficients of the mixture were then measured, at different concentration and temperature ranges. The resulting values of the Soret coefficient were plotted as a function of temperature and concentration. From the plots, a decrease in Soret coefficient was observed with decreasing temperature, and increasing toluene concentration. The data set was fitted with a second order polynomial and compared with results from previous investigations by Köhler and Müller [22], which showed excellent agreement. The results were also comparable to the results obtained by Ecennaro *et al.* [23], where a thermogravitational column technique was used. To check the validity of the results, a mixture of ethanol and water was also investigated. The resulting Soret coefficients were compared to values obtained by Kolodner *et al.* via a beam deflection technique, which also showed excellent agreement, and therefore validating the measured values. The diffusion coefficient for the toluene and n-hexane mixture was measured as well and plotted as a function of temperature and concentration. The results were compared with two other investigations where a 2% and 3% difference was seen. It was evident from the plots that the diffusion coefficient increased with temperature and decreased with toluene concentration, except for toluene fractions larger than 0.8.

Kita *et al.* [24] measured the thermal and Soret coefficients of a ternary mixture of polyethylene oxide (PEO) in mixed water-ethanol solvent over a wide range of solvent composition, by using the Thermal Diffusion Forced Rayleigh Scattering (TDFRS) method. This method involves creating an intensity grating by using the interference of two laser beams. A very small amount of dye is injected in the solution to absorb the energy and convert the intensity grating into a temperature grating. From the temperature grating, the corresponding composition and index of refraction gratings were determined. Further details regarding the TDFRS principle, possibilities and limitations are discussed by Wiegand *et al.* [25]. The quartz cell used in the experiment had a path length of 0.2 mm, and the defracted beam intensity was measured by a photomultiplier. The measurements were obtained at a room temperature of 22°C. Their results showed a sign change of the Soret coefficient occurs as the water content of the solvent increases. The sign change was observed at a water mass fraction value of 0.83 at room temperature. For higher water concentrations, the Soret coefficient is positive, meaning migration of the polymer to the cooler regions of the fluid, which is typical behaviour of polymers. On the other end, for lower concentrations of water, when the Soret coefficient is negative, the polymer is observed to move to the warmer regions of the fluid. Measurements were taken for two different polymer concentrations. At lower polymer concentration, a higher magnitude of Soret coefficient was observed. A water-rich polymer solution was also investigated with respect to the temperature dependence of the Soret coefficient. As temperature was increased, a sign change from negative to positive was observed. For validation of the TDFRS method, thermodiffusion experiments of water ethanol were also conducted and compared to values from previous literature data. For this binary mixture, a sign change of the Soret effect was observed at a water mass fraction value of 0.71, which shows great agreement with values obtained in other literature. In addition, a comparison of the thermodiffusion coefficient of the binary and ternary mixtures showed the

largest changes of the Soret coefficient occur in both cases in the concentration range. This is due to the structural changes of the hydrogen bonds, which affects the solubility of the polymer in water solutions.

Dutrieux *et al.* [26] measured the Soret coefficient using two independent methods for two different mixtures of water-ethanol, one containing 60.88 and the other 50 weight percent ethanol in water. The first method measured the Soret coefficient by using a 5-point sampling process in a thermogravitational column and measured the diffusion coefficient using the open-end capillary (OEC) technique. In the thermogravitational column, the liquid is situated in a vertical slot with a large aspect ratio, where a temperature gradient is applied to the two vertical walls made of copper material. The temperature was controlled by using a water thermal bath, with hot and cold mean temperatures of 25, 20 and 22.5°C, respectively. The dimensions of the cell used were 0.2 x 3 x 53 cm in the x, y and z-directions, respectively. Sampling was performed at five positions along the cell. The values of density as a function of vertical height showed a linear relationship, with density decreasing with cell height, because $S_T > 0$ and the lighter component migrates to the hot plate at the top of the column. The diffusion coefficients were measured using the OEC technique, where a small tube with one end open, filled with a binary solution with initial mass fraction c_0 , is immersed in a large volume bath with a mass fraction smaller than c_0 . The interaction – water diffuses outside the tube and alcohol inside the tube – is allowed until both mass fractions have reached an equilibrium state, and the diffusion coefficient is obtained. For this experiment, 32 tubes of 29mm height and 1 cm³ volume were filled with a solution with $c_0=0.6288$ in water and placed into 4 Plexiglas holders. Four vessels at the mean temperature of 22.5°C with a combined volume of 350 cm³ were connected to the thermal bath and were filled with the same solution with $c=0.5888$. The holders were then immersed in the vessels until equilibrium was reached. The diffusion coefficients measured were compared with other literature and showed a 3 to 4% difference.

with two investigators, but showed a 12% error with an earlier work by Kolodner *et al.*. The second method was a new technique developed by the authors. This method involves taking velocity field measurements of a transient state. The vertical velocity amplitudes are measured using Laser Doppler Velocimetry (LDV). Two mixtures were investigated as mentioned before. The velocity amplitude as a function of time for the first mixture, with a water mass fraction of 0.50, displayed an overshoot in the amplitude, whereas, in a plot of a pure substance such as pure ethanol, a steady amplitude is observed with no overshoot. Overall, the Soret coefficients obtained by the different methods were compared to each other, and the deviation between them was found to be between 3-4% which was deemed to be quite satisfactory by the authors. The results of the Soret coefficients for the $C=0.6088$ weight percentage of water at mean temperature were compared with other literature. The resulting Soret coefficient for this mixture was measured to be 3.23, which was in excellent agreement with other works. It was further concluded that reliable Soret coefficient values of organic liquid mixtures around room temperature could be obtained under terrestrial conditions.

Sapgnolo *et al.* [27] proposed an improved version of digital moiré to measure the isothermal diffusivity of liquid binary mixtures. The method uses a fringe generator with a diffractive optical element (DOE) illuminated by a spherical wave. The fringe pattern obtained is then projected on the bottom of a ground glass plate, and the cell is located in front of the glass. During the diffusion process, the grating pattern is recorded by a CCD camera and stored in the computer. The glass test cell dimensions were 20 x 8 x 75 mm. The light source was from a laser with constant wavelength projected onto a 'saw tooth' grating glass, which acts as a beam divider. The diffusion coefficients are obtained by using a proposed FFT method. To test the validity of this method, an experiment was carried out with a mixture of LiBr-water (1.0M) at 25°C. The phase difference was obtained and fitted with a polynomial fitting curve. The resulting

diffusion coefficient was $1.407 \times 10^{-5} \text{ cm}^2/\text{s}$, compared to a benchmark value of 1.404×10^{-5} which shows excellent agreement with the experimental value. Therefore, it was concluded that the improved method of digital moiré was a reliable technique to measure the diffusion coefficient for the mixture at room temperature, using the FFT method.

Mialdun and Shevtsova [28] measured the transport coefficients by using an experimental technique based on optical digital interferometry. This method differed from other similar optical techniques in that it is able to measure the temperature and concentration distributions along the diffusion path, as opposed to only measuring the concentration difference between a central point and a point. This allowed for examination of the convective flow inside the cell. The cell was a cubic transparent cell made of quartz glass, with length of 10mm. It was subjected to a temperature gradient from top (hot) and bottom (cold) by two copper plates, connected to a thermal bath, with temperature being controlled by Peltier modules. The unit was then placed in a setup based on the Mach-Zehnder interferometer concept. A light source from a laser with a constant frequency is expanded by a filter, then passes through a beam splitter which creates two rays of equal intensity where one of the beams passes through the cell. After passing through the cell and a mirror, the beam interferes with the reference beam at the second beam splitter. Finally the interference is captured by a CCD camera with a 1280×1024 pixel sensor. The entire setup was then placed on an optical table with pneumatic damping control to isolate external vibrations. The interference image is compared to an undisturbed one and the phase shift is determined by subjecting each image to a two-dimensional Fast Fourier Transform (2D FFT), then undergoing a filter of a selected region of the spectrum, then subjected to an inverse 2D FFT, and finally phase unwrapping. Initially, the cell was filled with a mixture of 50-50 weight percent where almost linear temperature profiles were observed near the horizontal walls and non-linear profiles for concentration were seen near the central part of the cell. However, moiré

away from the central regions, growing deviations were noticed, meaning convection was present in the cell. Therefore, a convection free zone was selected and validated by measuring the Soret and diffusion coefficients of water-ethanol mixtures with mass fractions of approximately 0.5 and 0.6. The measurements were taken at a mean temperature of 295.5 K and a temperature gradient of 9.87 K, with data collected at the central part of the cell using the fitting curve approach. Compared to data from other literature, the diffusion coefficient results as a function of water mass fraction were scattered among these data. This could have been due to the fact that different authors used different measurement methods to determine the diffusion coefficients. The obtained values in this work were below the cloud of data points from others. It was argued that if convection was present in the central region, it would cause an increase in the diffusion coefficient values and therefore it was concluded that this region was convection free. The Soret coefficients were calculated based on the fitting curve and using steady state, with values of 2.82×10^{-3} and 2.88×10^{-3} 1/K, respectively for a water mass fraction of 0.613. Compared with other data from the literature, via linear interpolation, a deviation of approximately 5% was observed. Therefore, it was concluded that the convection free zone in the cell was chosen correctly and that the optical interferometry technique provided good results. Measurements were also taken for a mixture of water-isopropanol, with mass fractions of 0.264, 0.5, 0.6 and 0.7. A comparison of the diffusion coefficient showed excellent agreement with other literature. The Soret coefficient values obtained for a temperature gradient of 10 K show excellent agreement with low concentration of water. As the water concentration is increased, divergence also increases in data agreement. Overall, it was concluded that the digital optical interferometry method is a reliable technique for transport coefficient measurements in terrestrial conditions.

Platten *et al.* [29] set out to measure and provide benchmark values of the Soret and diffusion coefficients of three binary mixtures of dodecane, isobutylbenzene (IBB) and 1,2,3,4

tetrahydronaphthalene (THN), for concentrations of 50 weight % at 25°C. These chemical mixtures are representative of the hydrocarbons found in oil reservoirs and therefore it is important to accurately determine the transport coefficients of these mixtures. Several different techniques were applied by five different research groups: transient holographic gratings, annular thermogravitational columns and vertical parallelepipedic columns with velocity amplitude determination by LDV. The first method mentioned will be discussed in more detail in the next paragraph. The second technique used a two-point sampling system in a thermogravitational column and determined the thermal diffusion coefficient from the concentration fraction difference between the top and bottom of the column. Knowledge of the thermal expansion coefficient and the kinematic viscosity was necessary. The last technique uses Laser Doppler velocimetry (LDV) to determine the time dependent velocity amplitude in order to obtain the influence of the Soret effect on the buoyancy force and to determine the separation ratio. The Soret coefficient is calculated by having knowledge of the thermal expansion coefficient and the mass expansion coefficient. By using a five-point sampling system, the value of the thermal diffusion coefficient is determined. Finally, using the open ended capillary (OC) technique, the diffusion coefficient is measured, and hence the ratio of D and D_T is compared with the value obtained in the LDV experiment. Overall, the differences between results from different laboratories were less than 7%, with the exception of one lab that produced a value for the thermal diffusion coefficient that showed an 8.5% difference. This large difference was attributed to the fact that columns with small gaps and larger gaps produced different results for unexplained reasons. Regarding the direct measurement of the Soret coefficient by the OC technique, for the mixture of THN and dodecane, there was found to be reasonable agreement between the direct method and the calculated value of the ratio of D_T and D . The final Soret coefficients measured for the mixtures THN-dodecane, THN-IBB and IBB-dodecane were

3.3 and 3.9×10^{-3} (1/K) respectively. These values, along with the obtained diffusion coefficients were considered to be benchmark values to be referred to for validation of method.

Wittko *et al.* [30] performed, as part of the bench mark experiments mentioned above in Platten *et al.* [29], a transient holographic grating technique to measure the Soret, thermal diffusion and mass diffusion coefficients of three binary mixtures of dodecane, IBB and THN, with mixtures of 50 weight % of each component at 25°C. The obtained data was analyzed using a new numerical correction algorithm based on a linear response theory. Measurements for the three mixtures were taken with five or more different laser intensities. The mean temperature of the sample is affected by higher laser intensity, meaning that at a high intensity, the mean sample temperature is higher than the temperature with low laser intensity, and therefore there was a slight difference in the coefficients. Therefore, the obtained results were fitted to a linear extrapolation to zero laser intensity to minimize the effect of the different sample temperatures. The Soret coefficients obtained for the mixtures of THN-dodecane, THN-IBB and IBB-dodecane were 9.45, 3.46 and 3.95×10^{-3} (1/K), respectively. Furthermore, experiments were performed to determine the thermal refractive index gradient and the refractive index gradient with respect to concentration with small relative errors.

From the various literature reviewed above, it is evident that many interferometric techniques exist, with results usually in good agreement, indicating that optical digital interferometry is a reliable way to determine the transport coefficients of liquids. In order to further support this statement, several experiments will be performed and compared to published benchmark results for comparison and further validation of the measurement method. The main comparison will be done with the benchmark values obtained by several researchers in the 'Fontainebleau' group. This group was started in 1999 in Fontainebleau, France, and consisted of five European

researchers, of which two have already been mentioned and discussed, namely Platten et al. and Wittko and Kohler [30]. This group measured the transport coefficients of binary mixtures of dodecane, IBB and THN, and after comparison and analysis, provided a set of benchmark data for these mixtures. Their methods of measurements differed from the present method of digital interferometry, and, therefore, a comparison between these sets of results will provide great insight into the accuracy and validity of measurements taken with optical interferometry.

1.2.3 Applications in Industry

The application of the Soret effect and thermodiffusion in general is widespread in different industries. For example, the Soret effect is present in biological systems, such as the transport induced by thermal gradients in biological cells. In order to determine to what extent biological systems could sustain conditions for thermal diffusion, Bonner [31] performed calculations based on the following models: a single living cell with the presence of heat removal at the surface by conduction, and the other model was a group of cells, for example an organ or tumor, again with the presence of heat removal at the surface by conduction. The performed calculations were meant to give a numerical basis to determine whether conditions could be sustained for thermal diffusion, and whether it could play a role in biological transport. The author concluded that for the models considered, thermal diffusion could indeed play a role in biological transport. It was determined that thermal diffusion could act as a regulator, and "lock in" or "lock out" certain substances specific to certain boundaries.

Another application is found in the oil industry, in which a lot of time and effort is currently invested to research thermal diffusion in crude oil. Currently, there is a lack of understanding of the characteristics of crude oil, making it therefore difficult to predict

conditions for natural hydrocarbon extraction. In order to understand the fluid behaviour at reservoir pressure and temperature conditions, the diffusion coefficient needs to be studied. These diffusion data are most important to the oil industry, because knowledge of this data is necessary to develop numerical models for oil reservoir characteristics. In order to attain data such as the thermodiffusion coefficient, it has been concluded that experiments must be conducted in a microgravity environment. The successful completion of such research allows oil companies to accurately predict the condition of a reservoir and dedicate the proper resources for extraction, resulting in higher overall efficiency. Foam injection, which is a method of oil recovery, also benefits from microgravity environment experiments, because the research techniques used to study foam are hindered by the gravitational forces, which mask other forces such as surface elasticity. Foam injection is used in many stages of oil recovery, such as well drilling, well production and reservoir injection. [32, 33]

1.3 Research Objective

Future IVIDIL (influence of vibrations on diffusion in liquids) experiments are planned on board the International Space Station (ISS). Since g-jitter is present on board the ISS, it is necessary to perform numerical analysis to get a better understanding of the effects caused on convection due to the presence of g-jitter. In order to perform this numerical study, data were obtained from experiments on board the FOTON M12 mission, which is similar to the g-jitter experienced on ISS. In addition, it is necessary to test the hardware that will be used for this future mission. Therefore, ground experiments are conducted to effectively determine the transport coefficients of several binary fluids. Therefore, the objective of this research is to determine the Soret coefficient of a binary mixture of water-isopropanol 90-10 weight % using a commercial finite element package. In addition, the Soret and thermal diffusion coefficients of binary mixtures of

dodecane, isobutylbenzene and 1,2,3,4-tetrahydronaphthalene are to be measured experimentally using digital optical interferometry, and to compare the results with benchmark values to establish the reliability of the digital interferometry technique. Finally, the Soret and diffusion coefficients of isopropanol – water will be determined experimentally, at different water fractions.

1.4 Thesis Organization

This thesis is organized in the following manner:

- In chapter one, a literature review of previous experiments and research concerning the techniques and results obtained in the measurement of transport coefficients of mixtures is covered.
- Chapter two discusses the governing equations used to obtain the results, for both numerical and experimental analysis. In addition, this section describes the boundary conditions and the solution techniques used in detail.
- Chapter three covers the experimental portion. It first covers a numerical analysis of the experimental setup, and then it explains the experimental Mach-Zehnder interferometry and the equipment properties. The experimental results obtained in terrestrial conditions are presented and validated by comparison with benchmark values.
- Chapter four discusses the numerical analysis performed. It explains the model in detail, presents results for the different scenarios examined, such as two-dimensional zero gravity and microgravity scenarios, and the three-dimensional g-jitter case.
- Chapter five gives a brief conclusion and future work.

Chapter 2. Numerical and Experimental

Formulation

2.1 Introduction

This chapter is divided into two sections, one for the numerical analysis part and the other for the experimental part of the thesis. The following sections will present the governing equations and the corresponding boundary conditions.

2.2 Governing equations used with the numerical part

The numerical and mathematical models include the three dimensional steady state Navier-Stokes momentum equations, the energy equation and the species equation. The governing equations correspond to the Boussinesq approximation and the Cartesian coordinate system. The Boussinesq approximation is often used in fluid dynamics, specifically buoyancy driven flows, and states that the differences in density are negligible, except when they are multiplied by acceleration due to gravity, 'g'. The equation used in the Boussinesq approximation will be presented in the following section. In addition to the mentioned governing equations, the theoretical equation used to solve the g-jitter case is presented and discussed.

2.2.1 Navier-Stokes Equations

The Navier-Stokes equations are a set of differential equations used to describe motion in fluids, such as gases or liquids. The principle of linear momentum in this study for the x, y and z directions in the Cartesian coordinates are as follows:

In general form:

$$\frac{D\vec{u}}{Dt} = -\frac{1}{\rho} \nabla p + \nu \nabla^2 \vec{u} + \vec{g} \quad (1)$$

In the x-direction:

$$\rho \left[\frac{\partial u}{\partial t} + u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y} + w \frac{\partial u}{\partial z} \right] = -\frac{\partial p}{\partial x} + \mu \left[\frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} + \frac{\partial^2 u}{\partial z^2} \right] + \rho g_x \quad (2)$$

In the y-direction:

$$\rho \left[\frac{\partial v}{\partial t} + u \frac{\partial v}{\partial x} + v \frac{\partial v}{\partial y} + w \frac{\partial v}{\partial z} \right] = -\frac{\partial p}{\partial y} + \mu \left[\frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial y^2} + \frac{\partial^2 v}{\partial z^2} \right] + \rho g_y \quad (3)$$

In the z-direction:

$$\rho \left[\frac{\partial w}{\partial t} + u \frac{\partial w}{\partial x} + v \frac{\partial w}{\partial y} + w \frac{\partial w}{\partial z} \right] = -\frac{\partial p}{\partial z} + \mu \left[\frac{\partial^2 w}{\partial x^2} + \frac{\partial^2 w}{\partial y^2} + \frac{\partial^2 w}{\partial z^2} \right] + \rho g_z \quad (4)$$

The above equations correspond to the Boussinesq approximation, where

$$\rho = \rho_0 [1 - \beta_T (T - T_0) + \beta_c (C - C_0)] \quad (5)$$

where T is the temperature and T_0 is the initial temperature at time zero seconds; ρ is the density; C is the mass concentration and C_0 is the initial concentration at time zero seconds; p is the pressure; u , v and w are the velocities in the x , y and z directions respectively; β_c and β_T are the volumetric concentration- and thermal expansion coefficients respectively.

In the case of the ideal condition (zero gravity), the last term in equations (1)-(4) is zero.

In the g-jitter case, the last term is calculated using:

$$g_x(t) = a_0 + \sum_{j=1}^m [a_j \cos(2\pi f_j t) + b_j \sin(2\pi f_j t)] \quad (6)$$

where a_0 , a_j and b_j are the Fourier coefficients and f_j is the frequency ($j = 1, m$). The equations for the y and z-direction are identical to equation (6). The above equation was used to determine the g-jitter results of an experiment on board the Russian FOTON-12 rocket. From this mission, the real Fourier coefficients and the on board frequencies were measured. Therefore, for this real scenario, the data and coefficients were used to model the three-dimensional case. The coefficients are presented in Table 2.2.

2.2.2 Energy Equation

The energy equation is used to describe the change of temperature in the model. Assuming constant thermal conductivity and no internal heat generation in the model, the principal of conservation of thermal energy is as follows:

$$\frac{DT}{Dt} = \frac{k}{\rho c_p} \nabla^2 T \quad (7)$$

where c_p is the specific heat and k is the thermal conductivity. The energy equation can be expanded to

$$\left[\frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x} + v \frac{\partial T}{\partial y} + w \frac{\partial T}{\partial z} \right] = \frac{k}{\rho c_p} \left[\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} \right] \quad (8)$$

2.2.3 Species Conservation Equation

The species equation is used to describe the concentration change and is as follows:

$$\frac{DC}{Dt} = D_c \nabla^2 C + D_T \nabla^2 T \quad (9)$$

where D_c and D_T are the molecular and thermal diffusion coefficients respectively, and ρ is constant in order to see solely the effects of g-jitter on the model. Equation (9) can be expressed

$$\text{to } \frac{\partial}{\partial t}(\rho c_i) + \nabla \cdot (\rho \vec{u} c_i) = -\nabla \cdot \vec{j}_i, \quad (i = 1, 2), \text{ where the diffusion flux } \vec{j}_i = -\rho \left(\sum_{j=1}^2 D_{ij} \nabla c_j + D_{T,i} \nabla T \right) \\ = 1, 2)$$

2.3 Boundary and Initial Conditions

It is assumed that no reactions occur inside the cavity and no mass flux through the walls. The flow and no slip boundary conditions are applied to the side walls, which are also assumed adiabatic. The three-dimensional experimental cell as modeled in the commercial finite element software COMSOL 3.4 can be seen in Figure 2.1, which shows all the interior and exterior edges.

Figure 2.1: Three-dimensional model (edge mode)

The remaining conditions are given by:

At $x=0, x=L$, where L is the full length of the model at $x = 0.04$ [m] (see Figure 2.1)

$$u=0$$

(10)

$$T_{x=0}=T_{\text{cold}} \quad (11)$$

$$T_{x=L}=T_{\text{hot}} \quad (12)$$

$$J_x = D_c \frac{\partial C}{\partial x} + D_T \frac{\partial T}{\partial x} = 0 \quad (13)$$

At $y=0, y=W$, where W is the full width of the model at $y = 0.012$ [m]

$$v=0 \quad (14)$$

$$J_y = D_c \frac{\partial C}{\partial y} + D_T \frac{\partial T}{\partial y} = 0 \quad (15)$$

$$\frac{\partial T}{\partial y} = 0 \quad (16)$$

At $z=0, z=H$, where H is the full height of the model at $H = 0.012$ [m]

$$w=0 \quad (17)$$

$$J_z = D_c \frac{\partial C}{\partial z} + D_T \frac{\partial T}{\partial z} = 0 \quad (18)$$

$$\frac{\partial T}{\partial z} = 0 \quad (19)$$

where L, H and W are length, height and width respectively of the cavity; J_x, J_y and J_z are the diffusion fluxes in the X-, Y- and Z-directions.

The initial conditions at time $t=0$ are

$$\vec{u} = 0, C_0=0.9, p_0=0, T_0=298\text{K}.$$

2.4 Solution technique and convergence criteria

The numerical procedure of this experiment involves using the commercial finite element package COMSOL Multiphysics 3.4 to solve for the governing equations, along with the described boundary conditions.

The equations are solved for each mesh point in the cavity. The energy, incompressible Navier-Stokes momentum equations and species equations are built in the software, and can be found in the 'Heat Transfer', 'Fluid Dynamics' and 'PDE Modes – General Form' sections in the 'Chemical Engineering' module, respectively. All modes required parameters of the mixture to be input by the user. For a list of the thermo-physical properties of water-isopropanol, refer to Table 2.1.

Table 2.1: Thermo-physical properties of water-isopropanol (90-10 weight %)

ρ_0	ν	μ	c_p	κ	β_T	β_c	D_c	S
984.000 (kg/m ³)	1.410 x 10 ⁻⁶ (m ² /s)	1.387 x 10 ⁻³ (Pa s)	3990.500 (J/kg K)	0.522 (W/m K)	3.100 x 10 ⁻⁴ (1/K)	0.1386	8.700 x 10 ⁻¹⁰ (m ² /s)	-10. 1 (1)

For the incompressible N-S mode, an acceleration factor was added to the domain settings, with a tuning factor of 0.25. This option was utilized because it allowed for increased stability in the system. The cell wall boundaries adopt the no-slip condition.

The linear solver used to solve the problem was the 'UMFPACK' direct solver, written by Timothy A. Davis¹, which solves general systems of the form $Ax = b$ using the 'multifrontal' method and direct LU decomposition of the sparse matrix. The multifrontal method is a variation of the Gauss elimination method and is used to solve sparse linear systems. This method avoids

¹ Timothy A. Davis, <http://www.cise.ufl.edu/research/sparse/umfpack>

large number of operations involving zero terms and builds 'LU' of a sparse matrix (LU, a matrix decomposition, writes a matrix as the product of a Lower and Upper triangular matrix). Since UMFPACK was used as a direct solver, no options existed in the solver parameter settings to choose a preconditioner (a preconditioner is useful in iterative methods used to solve large sparse linear systems).

Manual tuning of the initial time-step was selected to be 10^{-9} in order to allow the solution to converge at initial time-step. The default value of the software would result in divergence of the solution. This feature was only used for the initial time-steps, and was turned off for further steps. The time step was chosen carefully to allow for the different scenarios in order for the solution to run error free (divergence), and to produce the required data in a reasonable time. If the time step chosen is too small, then the computer could run out of memory to process the necessary calculations and data storage, or the computation would be very long in duration. The appropriate time steps chosen for the different scenarios will be included in the COMSOL model report in Appendix A.

The solver continues operation until convergence is achieved for each time-step. This state is achieved when the maximum relative error between two successive iterations is less than 10^{-6} .

A custom mesh was generated in order to achieve convergence throughout the model, especially near the boundaries where high instabilities could cause divergence. Therefore the mesh was made extra fine near the boundaries.

In order to avoid a temperature discontinuity at the boundaries, a 'smoothed step function' was introduced as the initial condition for the temperature equation. The role of the step function is to enhance the numerical stability and convergence of the model. It uses a smoothed function with a

continuous second derivative, and approximates the function by smoothing the transition within the specified interval scale.

In addition to the above settings implemented in the software to allow the model to be solved with ease, the initial pressure was also set to zero rather than atmospheric pressure. This allowed more stability in the model.

In the case of g-jitter analysis, data from Foton-TRAMP was stored in the constants subdomain expressions in COMSOL. The Foton-TRAMP data is listed in Table 2.2:

Table 2.2: Fourier coefficients for Foton-TRAMP [9]

X-direction				
j	a_0 (10^{-6} m/s ²)	a_j (10^{-6} m/s ²)	b_j (10^{-6} m/s ²)	f_j (10^{-3} Hz)
1	-3.6615	-24.896	30.786	1.645209
Y-direction				
j	a_0 (10^{-6} m/s ²)	a_j (10^{-6} m/s ²)	b_j (10^{-6} m/s ²)	f_j (10^{-3} Hz)
1	-116.65	1.5392	0.42332	1.157556
2	--	3.3976	-7.5033	2.51535
3	--	4.1744	1.4193	1.872925
4	--	-4.7073	-2.6550	2.237846
5	--	-0.96563	-1.4247	3.281500
Z-direction				
j	a_0 (10^{-6} m/s ²)	a_j (10^{-6} m/s ²)	b_j (10^{-6} m/s ²)	f_j (10^{-3} Hz)
1	16.508	5.5583	2.8422	1.643534
2	--	-0.45562	-0.70390	1.870828
3	--	-1.5791	-0.094072	2.073682
4	--	-2.8356	1.0229	3.290399
5	--	-0.91597	-0.27967	4.149568

After computations were made, it was possible to plot desired variables directly in COMSOL itself, via its post processing options, or to export the data and use other software, such as Microsoft Excel to further analyze data. Further details on the parameters and settings used in COMSOL can be found in model report in Appendix A

2.5 COMSOL Multiphysics

The commercial software COMSOL is a powerful simulation environment which allows the user to conduct a wide range of modeling tasks, such as defining geometry, implementation of boundary and sub-domain settings, meshing, solving and post-processing analysis of results. Creating models in this platform is easy and quick when compared to other similar commercially available packages. This is mainly due to the high emphasis of Graphics User Interface (GUI) functions, making the software very user friendly. Incorporated in the software are previously generated models for a wide variety of applications, ranging from heat transfer and fluid dynamics to structural mechanics.

Its flexibility becomes apparent when additional physics templates are added and implemented with ease. Also, options such as the ability to implement equations into the model with ease make the software more practical and efficient.

Depending on the complexity of the problem, solutions and simulations are achieved fast. The post processing sections allows for a variety of analysis options, with the ability to create high quality plots of the results.

2.6 Governing equations for the experimental part

The governing equations for motionless fluids are applied, which are the Energy and the Species equations (equations 7 and 9) already mentioned in section 2.1.2 and 2.1.3. The optical method relies on the variation of refractive index with composition. The below equations are used to determine the change in refractive index, from which variations in temperature and concentration are obtained.

2.6.1 Phase change

In order to determine the gradients of refractive index, it is first necessary to determine the shifts in the x and z directions, determined by:

$$\Delta\varphi(x, z, t) = \Delta\varphi_{liquid}(x, z, t) + 2\Delta\varphi_{cell}(z, t) \quad (20)$$

2.6.2 Change of refractive index

Having knowledge of the unwrapped phase change, the refractive index can be calculated using

$$\Delta n = n(x, z) - n_0 = \frac{\lambda}{2\pi L} \Delta\varphi \quad (21)$$

where λ is the wavelength and L is the thickness of the liquid in the direction of the optical pathway.

2.6.3 Fitting curve equations

For the analysis of the results for the diffusion curve, the following fitting curve equation was used to determine the concentration difference between two convection free zones, z_1 and z_2 :

$$\Delta C(z_2 - z_1, t) = -S_T C_0 (1 - C_0) \Delta T \left[\frac{\Delta z}{L} - \frac{4}{\pi^2} \sum_{n, odd} \frac{1}{n^2} \exp\left(-n^2 \frac{t}{\tau_r}\right) \left[\cos\left(\frac{n\pi z_1}{L}\right) - \cos\left(\frac{n\pi z_2}{L}\right) \right] \right] \quad (22)$$

The two unknown parameters in the above equation are the Soret coefficient S_T and the diffusion

relaxation time $\tau_r = \frac{L^2}{\pi^2 D}$

Another equation used for the diffusion fitting curve [28] was

$$\Delta C(z_2 - z_1, t) = -\Delta C_0 \frac{4}{\pi^2} \sum_{n, odd} \frac{1}{n^2} \exp\left(-n^2 \frac{t}{\tau_r}\right) \left[\cos\left(\frac{n\pi z_2}{L}\right) - \cos\left(\frac{n\pi z_1}{L}\right) \right] \quad (23)$$

where the unknown parameter is the diffusion relaxation time τ_r .

2.6.4 Two-dimensional Fast Fourier Transform

The Fast Fourier Transform (FFT) is a more efficient algorithm developed in 1965 by Cooley and Tukey [34] to solve the Discrete Fourier Transform (DFT), which is a specific form of Fourier analysis. Essentially, the DFT performs a linear transformation of one function into another, in the frequency domain. This transformation is represented by the below equation, which is for the two-dimensional case:

$$F(u, v) = \frac{1}{MN} \sum_{x=0}^M \sum_{y=0}^N f(x, y) e^{-i2\pi \left(\frac{ux}{M} + \frac{vy}{N} \right)} \quad \text{for } n = 0 \dots N-1 \quad (24)$$

where $f(x, y)$ represents the complex amplitude of the input signal, and $F(u, v)$ is the Fourier transform.

The FFT is called fast, because this algorithm reduces the computational time for M points from $2M^2$ to $2M \log_2 M$.

Chapter 3. Experimental Measurement of Soret

Coefficients for Liquid Mixtures

3.1 Experimental Setup and equipment properties

Experiments were performed at the Microgravity Research Centre (MRC) at the Universite I de Bruxelles in Brussels, Belgium. The experimental setup is based on a Mach-Zehnder interferometer concept, shown in Figure 3.1 below:

Figure 3.1: Schematic of experimental setup

The light source (ThorLabs Inc. USA.) used in the experiment was a constant frequency He-Ne laser, with wavelength $\lambda = 632.8$ nm. The optical path of the beam in the experimental liquid was $L = 0.018$ m. The incident beam first passes through a spatial filter (Melles Griot) where it is expanded, then passes through the first beam splitter, where it is split into two beams with equal intensity. One beam is deflected to a mirror and bypasses the cell, whereas the other beam travels through the cell. Once both object and reference beams are deflected by mirrors, they interfere with each other at the second beam splitter, and the interference fringes are captured by a CCD camera (Progressive scan: JAI Corp. Japan, lens: Nikon, Japan) with a 1280×1024 pixel sensor. In order to adjust and modify the distance between the fringes by changing the incident angles of the object and reference beams, it is possible to change the inclination angle of one of the mirrors (ThorLabs Inc. USA). The CCD camera is connected to a computer, where the images are stored. The actual setup can be seen in Figure 3.2.

Figure 3.2: Optical experimental setup based on Mach-Zehnder interferometry

In the above picture, the laser is the long horizontal tube at the top. On the right side, arranged vertically, is the sequence of beam expander, mirrors and beam splitter. On the left side are mirrors and the beam splitter, and finally the CCD camera.

The cell itself is made of quartz Suprasil by Helma Company, with outer dimensions of [20 x 6.2 mm]. A technical drawing of the cell is available in Appendix B.

A SolidWorks drawing of the experimental cell can be seen in the Figure 3.3.

Figure 3.3: SolidWorks drawing of cell, with copper plates installed

From the above figure, one can see the experimental cell is located in the centre between copper plates. The hole in the bottom plate is the mixture feed and drain path where the tubes are connected. The top hole is used to allow air to travel in or out of the cell when draining or feeding. The plate below the cell is connected to another copper plate, which is used for water feed from the thermal bath, hence the two pipes. In between the two plates is the P

module to control the cell temperature. This same configuration is repeated for the top portion, which can be seen in the picture below, taken of the actual cell setup:

Figure 3.4: Picture of the actual experimental cell and hardware.

It should be mentioned that two different sealants were used during the experiments between the glass cell and the copper plates. For experiments involving the Fontainebleau mixtures (which will be explained in section 3.3), a PTFE (Polytetrafluoroethylene or Teflon) based sealant was necessary, due to the corrosive properties of the chemicals used when in contact with rubber compounds. Therefore, in order to avoid leakage, it was decided to use a Teflon based sealant that would not dissolve during the experimentation. It was suspected though, that the properties of the sealant did affect convection and mixing in the fluid during thermodiffusion. This effect

was less pronounced and noticeable when a regular rubber sealant was used for experiments involving water-isopropanol mixtures.

As mentioned, the temperature of the copper plates above and below the cell were controlled by Peltier modules driven by electronic temperature controllers. The entire experiment setup was mounted on a plate. This plate was mounted on a Melles Griot table with damping control in order to minimize external vibrations.

The chemicals used for the experiment were from Acro Organics and are as follows: Isobutylbenzene (IBB) 99.50% purity, 1,2,3,4 – tetrahydronaphthalene (THN) 98+% purity, dodecane 98% purity, and ethanol. For more information on the liquids, refer to Appendix C.

3.2 Experimental procedure

3.2.1. Experimental preparation

Before the actual experiment was started, several thermistors were calibrated by being placed in an aluminum block that was then placed in a thermal calibration bath (Haake F3, Germany) at 20°C. The temperature was increased to 30°C at increments of +2°C with at least 15 minutes between each interval. The data was acquired by the Agilent data acquisition unit model 34970A. With the thermistors calibrated the experiment could be started. The experimental cell was filled with a binary mixture, which was made using a sensitive scale (Sartorius, Germany) and a cylindrical glass vessel with weight 16.695 g. The filling procedure was a delicate task, as no air or bubbles were to be present in the system. Therefore, a de-gassing unit (Systec OEM Model 100 Vacuum Degasser, USA) was used to ensure the system was vacuum and free of any residual gases. The insertion was performed by using a glass syringe connected to very narrow diameter PTFE lined tubes. The insertion tube was connected to the bottom of the cell, and filled to

top, where another tube was connected for discharge. This procedure of filling and discharging was repeated at least two or three times to ensure any residual liquid from previous experiments were not present in the system. The insertion itself was done by placing the syringe in a programmable pump machine (Harvard Apparatus PHD 2000, USA) and setting refill parameters such as a refill rate of 0.1 mL/min, target volume of 3.5 mL, and a syringe diameter of 10.3 mm. Once the cell was filled successfully, it was placed in the experimental structure. The experimental structure itself was located in a thermal insulated box in order to control the temperature surrounding the experimental cell.

The experimental setup for filling the cell can be seen in Figure 3.5. At the top of the picture is the pump with the syringe secured. In the middle are the de-gasser, and finally the cell. As mentioned, very thin tubes were used in this process, as seen in the picture.

Before data acquisition of images were started, it was necessary to allow the temperature of the thermal box around the apparatus as well as the copper plates to reach a steady temperature, set to 25°C. Three thermistors were involved in the exterior temperature control. Two of the thermistors measured temperatures in the box: one by the control fan module, the other on the optical plate of the experimental setup. The third was placed outside the box and measured the temperature of the dampening table on which the experiment was placed. At the same time, some images were obtained during this stage. After about one hour, the Peltier modules were switched on and allowed to match the temperature of the cell with the plates and surrounding air. This step also allowed the temperature of the modules to reach a steady state. This step would usually take about 2 hours. Once a desired steady state was reached, a phase image could be chosen to use as the reference phase. At this point, a temperature gradient of 6°C was applied to the cell, and data

Figure 3.5: Experimental setup showing cell filling arrangement and apparatus

acquisition would record images at specified intervals. Initially, the CCD camera would ob
images every minute, and then gradually this time step would be increased as thermodiffus
became slower. The experiment would run for about 20-24 hours until the temperature grad

was removed. At this point, image acquisition would continue to for the relaxation phase, which would also take about 20-24 hours.

3.2.2. Processing of interferogram

A sample of a typical interferogram obtained from the experiment is shown below:

Figure 3.6: Typical (cropped) raw interferogram obtained during experiment data acquisition

As mentioned earlier, the laser beam is split in two, where one travels through the cell and the other bypasses the cell and remains undisturbed. These two beams then coincide and produce and interference, which produces a set of dark and light lines, called fringes, as shown in the above picture. This result is a raw image and needs to be analyzed and translated into something more sensible. Therefore the images are reconstructed by performing a two dimensional Fast Fourier Transform (FFT). The theoretical equations for this method were mentioned in Chapter 2.

There were different segments of data acquisition in the experiment procedure. First, the insulated box temperature would be set to a room temperature of 25°C, and allowed to reach a steady state, which would typically take a couple of hours. The next step would be to introduce the room temperature to the experimental cell, via the Peltier modules. Once a steady state was reached, the temperature gradient was introduced to the cell, and data acquisition continued. The

time for thermal diffusion to reach a steady state was typically about 20 hours. After this p the temperature gradient would be removed, and the experimental cell would be set to temperature once again, to allow for thermal relaxation. This would take about 15 hours.

From the images obtained in the first segment, for the steady state, one image would be selected to be used as a reference image, to determine the temperature distribution once a temperature gradient is applied. Typically, the image selected would be the last image recorded just before the temperature gradient is applied. When the temperature gradient is applied, the time step for the image acquisition is set to record an image per minute. This time step would be kept at about half hour, and gradually increased to track the slower thermodiffusion separation. The sequence of image processing is depicted in Figure 3.7:

Figure 3.7: Principle of the image processing procedure using the two-dimensional FFT[31]

First, the raw image is resized to a desired pixel size, $2^m \times 2^n$. The resized image is then transformed into the Fourier domain by applying the two-dimensional fast Fourier transform, followed by a filtering process. An inverse two-dimensional FFT is then applied, followed by phase unwrapping.

The mathematical codes developed to analyze and translate the fringe images into something more sensible were written and executed in MATLAB v. 7.1. From the reference image, several scripts, previously written by A. Mialdun² (Refer to Appendix D for code), are used to determine the temperature distribution.

²

A. Mialdun. Université Libre de Bruxelles – MRC, CP-165/62. 50 av. F.D. Roosevelt, B-1050 Brussels, Belgium

the desired region in the image to process, and apply the two-dimensional Fast Fourier Transform (FFT) for phase extraction and phase unwrapping. The unwrapped phase array is then filtered to exclude distorted edges.

It is necessary to note that during the process of reference image determination, the fringe image area to be used is determined manually and its corresponding coordinates are entered in the source code. In addition, the appropriate parameters and characteristics of different mixtures, initial concentrations, etc. need to be specified for each mixture on a case-by-case basis. The first reference image, corresponding to the undisturbed, homogenous state of the mixture, is determined by first choosing an acceptable region on the image, then plotting the phase differences in both the x and z directions observed with time up to the point when a temperature gradient is applied. The most stable point on this plot is picked and the corresponding image is picked as the reference image. An example can be seen in Figure 3.8.

The refractive index variation with temperature is then calculated and plotted for each image. From this resulting plot, another reference image is selected at the point where there is a steady variation of the refractive index. This image is then used as a reference image to determine the change in refractive index due to concentration change. The images selected for this section are the images recorded when the temperature gradient is applied to the experimental cell. The change in refractive index and change in concentration is determined following the same procedure described above. Once the change in concentration is calculated and plotted versus time, a fitting curve, equation (22) is applied, and the diffusion, Soret coefficients and the relaxation time are determined.

Figure 3.8: Phase difference plots in the X and Z directions vs. time. The circled data point is selected as the reference image

Again, it should be noted that a ‘correction’ procedure was implemented to determine diffusion and Soret coefficients for the thermodiffusion process, involving the selection of appropriate layer thickness. In the concentration plots, some perturbations or small convective

were observed near the top and bottom walls, as well as the corners. Therefore, in order to determine the diffusion coefficient, it was necessary to choose a region for concentration where there were no presence of perturbations. As can be seen in Figure 3.9, the centre of the field does not experience any disturbances in the concentration distribution.

Figure 3.9: Concentration distribution of IBB-THN. Small Perturbations visible in all four corners

The appropriate layer thickness is then selected by plotting the variation of the Soret and diffusion coefficients against variation of layer thickness. The point where a stable variation is observed is picked, and the corresponding layer thickness is used to determine the Soret and diffusion coefficients. An example can be seen in Figure 3.10, in this case for a 50-50 wt% mixture of water-ethanol.

Figure 3.10: Plots of Standard concentration deviation, diffusion, Soret and initial concentration change vs. layer thickness, respectively

The images corresponding to the relaxation period, when the temperature gradient is removed, are also processed in the same manner. Another fitting curve is applied to this set of images and equation (23) to determine the diffusion coefficient. This equation contains two parameters that must be known: one is the Soret coefficient, and the other is the diffusion relaxation time. The Soret coefficient is obtained in the transient state of the experiment, and the diffusion relaxation

time is the time from removal of the temperature gradient to when relaxation has reached a steady state.

The two diffusion coefficients calculated are then combined, and a final mean value is presented for comparison with other literature and benchmark values.

3.3 Results and discussion

Using the experimental setup of digital interferometry described above, experiments were performed to determine the diffusion and Soret coefficients of several binary mixtures. The resulting data were then analysed and plotted using a fitting curve, as described in Chapter 2.

The mixtures used in the digital interferometry experiments are water-isopropanol, IBB-dodecane, IBB-THN, THN-dodecane and water-ethanol, all with equal mass fractions of 50 weight percentage. Other mixtures used are water 90 wt%-ethanol and dodecane 60 wt%-THN.

The experimental Soret and diffusion coefficients are presented and compared to previous published results from various authors in the following sections.

3.3.1. Method validation against Fontainebleau benchmark

In 1999 in Fontainebleau, France, the binary mixtures of Dodecane, IBB and THN were chosen to be investigated by five European researchers to determine the transport coefficients of these mixtures before they were tested on the International Space Station. Also, these mixtures were chosen because the amount of data available on the Soret and diffusion coefficients of these mixtures was small. The experiments each had different methods of measurement, which will not be discussed in this thesis, as they are discussed in Ref. 32 and Ref. 33. Their individual results are presented in Table 3.1, and their combined, benchmark values are presented in Table 3.2. The present results of this thesis are compared to these benchmark values.

Table 3.1: Individual results for Fontainebleau binary mixtures ($T_0=25^\circ\text{C}$, $c_0=0.5$), Platten et al. [32]

Mixture	D ($10^{-10} \text{ m}^2/\text{s}$)				S _T (10^{-3} K^{-1})		
	Univ. Mons-Hainaut	Univ. Bayreuth	Max-Planck Inst.	Univ. Mons-Hainaut	Univ. del Pais Vasco	Univ. Bayreuth	Max-Planck Inst.
THN-C ₁₂ H ₂₆	6.15	6.26	6.22	10.20	8.73	9.45	9.52
THN-IBB	9.05	8.4	7.9	3.14	3.03	3.46	3.55
IBB-C ₁₂ H ₂₆	9.21	9.9	9.32	3.69	3.96	3.95	3.95

Table 3.2: Proposed benchmark values for Fontainebleau mixtures, Platten et al. [32]

Mixture	D ($10^{-10} \text{ m}^2/\text{s}$)	S _T (10^{-3} K^{-1})
THN-C ₁₂ H ₂₆	6.21 ± 0.06	9.5 ± 0.5
THN-IBB	8.5 ± 0.6	3.3 ± 0.3
IBB-C ₁₂ H ₂₆	9.5 ± 0.4	3.9 ± 0.1

3.3.1.1 IBB-C₁₂H₂₆ Mixture

The results for IBB-C₁₂H₂₆ (Dodecane) are presented in Figures 3.11 and 3.12. Figure 3.11 shows the concentration change over time, with the resulting Soret and diffusion coefficient $S_T = 3.98 \times 10^{-3} \text{ 1/K}$, and $D = 9.40 \times 10^{-10} \text{ m}^2/\text{s}$. Figure 3.12 presents the concentration change over diffusion relaxation time, when the temperature gradient is removed, and the mixture is allowed to return to the initial state at $T_0 = 25^\circ\text{C}$. The resulting diffusion coefficient is $D = 9.23 \times 10^{-10} \text{ m}^2/\text{s}$. These values correspond to a chosen layer thickness of $\Delta z = 3.4 \text{ mm}$. The procedure for layer thickness determination has been described in a previous section.

Compared to the benchmark values, the results are in very good agreement and fall well within the ‘safe’ zone, meaning the acceptable error range of the benchmark values, as depicted in Figure 3.13 and 3.14.

Figure 3.11: Thermodiffusion plot of concentration difference vs time for IBB-50wt%Dodecane

Figure 3.12: Diffusion plot of concentration difference vs time for IBB-50wt% Dodecane

Figure 3.13: Comparison of present experimental diffusion values with other literature results for Dodecane-50wt%IBB

Figure 3.14: Comparison of present experimental Soret values with other literature results for Dodecane-50wt%IBB

3.3.1.2 IBB-THN Mixture

The results for IBB-THN are in even better agreement with the benchmark values compared to the previous mixture. The resulting coefficients, as presented in Figure 3.15, are $S_T = 3.29 \times 10^{-3} \text{ K}^{-1}$ and $D = 8.6 \times 10^{-10} \text{ m}^2/\text{s}$.

The same can be said for the diffusion coefficient from the relaxation portion of the experiment, as presented in Figure 3.16, with a diffusion coefficient of $D = 8.43 \times 10^{-10} \text{ m}^2/\text{s}$. These values correspond to a layer thickness of $\Delta z = 3.1$ and 3.4 mm , respectively.

Figure 3.15: Thermodiffusion plot of concentration difference vs. time of IBB-50wt% THN

Figure 3.16: Diffusion plot of concentration difference vs time of IBB-50wt%THN

The excellent agreement of the results is once more compared to other individual data obtained via different methods and presented in Figures 3.17 and 3.18.

Figure 3.17: Comparison of present experimental diffusion values with other literature results for IBB-50wt%

Figure 3.18: Comparison of present experimental Soret values with other literature results for IBB-50wt% THN

3.3.1.3 THN- $C_{12}H_{26}$ (Dodecane) Mixture

The present results of THN- $C_{12}H_{26}$ are shown in Figure 3.19. The coefficients obtained during the thermodiffusion stage of the experiment are $S_T = 8.72 \times 10^{-3} \text{ K}^{-1}$ and $D = 6.27 \times 10^{-10} \text{ m}^2/\text{s}$.

Figure 3.19: Thermodiffusion plot of concentration difference vs time of Dodecane-50wt% THN

These values correspond to a layer thickness of $\Delta z = 4.9$ mm. The diffusion coefficient for diffusion relaxation segment is $D = 6.37 \times 10^{-10} \text{ m}^2/\text{s}$, as shown in Figure 3.20, which also shows the evolution of concentration change with respect to time. For this experiment, the results are acceptable. With an average diffusion coefficient value of $D = 6.32 \times 10^{-10} \text{ m}^2/\text{s}$, both diffusion and Soret coefficients fall just outside of the error margin when compared to benchmark values, as shown in Figures 3.21 and 3.22.

Figure 3.20: Diffusion plot of concentration difference vs time of Dodecane-50wt% THN

Figure 3.21: Comparison of present experimental diffusion values with other literature results for Dodecane-50wt% THN

Figure 3.22: Comparison of present experimental Soret values with other literature results for Dodecane-50wt%THN

3.3.1.4 Dodecane-THN Mixture

The 50-50 wt% mixtures of IBB, THN and Dodecane from the above sections showed very good agreement when compared to the benchmark values. However, no known data measurement exists for a mixture of Dodecane – 40wt% THN (Dodecane: 11.9987 g, THN: 8.0041 g). In order to obtain further knowledge about the behaviour of the transport coefficients of this mixture away from the usual 50-50 wt% composition, it was decided to perform the digital interferometry experiment with a 60-40 mass fraction. In this case, there are no other published results to compare to the present experimental result. The contrast factor of the mixture was calculated by using a second order polynomial from Wittko et al. [33], $dn/dc = 11.02 \times 10^{-4}$ 1/wt%.

The results for thermodiffusion are shown in Figure 3.23, which also shows the evolution of concentration change data with time, as well as the fitting curve. The concentration change reaches the steady state after about ten hours. For the determined layer thickness of $\Delta z = 5.7 \times 10^{-4}$

³ m, the resulting Soret coefficient is $S_T = 7.57 \times 10^{-3} \text{ 1/K}$, and the diffusion coefficient $D = 6.70 \times 10^{-10} \text{ m}^2/\text{s}$.

Figure 3.23: Concentration change vs. thermodiffusion time for dodecane-40 wt% THN

Once the temperature gradient was removed, the diffusion relaxation produced a diffusion coefficient of $D = 6.88 \times 10^{-10} \text{ m}^2/\text{s}$, Figure 3.24, for a layer thickness of $\Delta z = 5.6 \times 10^{-3} \text{ m}$, slightly larger than the previous thermodiffusion layer thickness value. Again, there are no similar data to compare to. However, compared to the benchmark value for a 50 wt% composition, $D = 6.21 \times 10^{-10} \text{ m}^2/\text{s}$, it is evident that the diffusion coefficient is increased slightly. The opposite holds true for the Soret coefficient, which shows a decrease, compared to the benchmark value $S_T = 9.5 \times 10^{-3} \text{ 1/K}$.

In order to fully understand the behaviour of the transport coefficients for this binary system, it is necessary to conduct further experimentation at a full range of different compositions. This work will be left for further research.

Figure 3.24: Concentration change vs. diffusion relaxation time for dodecane-40 wt% THN

3.3.2. Additional validation with ethanol-water mixtures

In addition to performing the above mentioned Fontainebleau experiments for validation of the method, another experiment was performed to further support the validity and provide greater confidence in the results. The mixture chosen to validate the results was a binary mixture of water 50 wt%-ethanol. This particular mixture was chosen due to the large amount of data for measurements of the Soret and diffusion coefficients available for this mixture. The mixture tested contained 9.4978 g of distilled water, and 9.4958 g of ethanol. It should be noted that due to different saturation values of the vapour pressure for water and ethanol, the composition changes during the degassing process, usually to the amount of approximately 2% [31]. The initial temperature of the experiment was set to 25°C, as with the other experiments, with initial concentration $C_0 = 0.5$. The resulting change in concentration over time can be seen in Figure 3.25. The curve in the plot is obtained by applying the fitting curve, equation (22), presented in

Chapter 2. Note that the results and curve correspond to the centre area, where there is convection present, with a height of $\Delta z = 4.9 \times 10^{-3}$ m, and $\Delta T = 5.7$ K. The resulting S_T coefficient is $S_T = 4.16 \times 10^{-3} \text{ K}^{-1}$, and the diffusion coefficient $D = 3.16 \times 10^{-10} \text{ m}^2/\text{s}$. The diffusion coefficient obtained from the relaxation stage is $D = 3.56 \times 10^{-10} \text{ m}^2/\text{s}$, as seen in Figure 3.26. Taking the mean value of the diffusion coefficients results in a mean value of $D_{\text{mean}} = 3.36 \times 10^{-10} \text{ m}^2/\text{s}$.

Figure 3.25: Concentration change vs time of water-50 wt%ethanol

To determine the validity of these results, it is necessary to compare them to other literature results. The literature results were obtained using different methods, such as thermal diffusion

Figure 3.26: Diffusion plot of concentration difference vs time of Ethanol-50wt% water

forced Rayleigh scattering (TDFRS) method used by Kita et al. [27], open-ended capillary method by Dutrieux et al. [29], diaphragm cell technique by Hammond and Stokes [35], Taylor dispersion method by Harris et al. [36] and Pratt and Wakeham [37], and digital interferometry by Mialdun and Shevtsova [28] as seen in Figure 3.27:

As evident from the above plot, the mean value of the diffusion coefficient at 0.5 water mass fraction obtained from present results is in excellent agreement with the previous results of Mialdun and Sevtsova, and Hammond and Stokes. Furthermore, the result is below the scatter of results from Dutrieux et al. and Kita et al. Should convection be present, it would cause an increase in the diffusion coefficient value. Therefore, it can be concluded that the obtained diffusion value is valid. Furthermore, it shows that the layer thickness (Δz) chosen for the data

Figure 3.27: Plot of various diffusion coefficients vs. different water-ethanol mass fractions

analysis is correct, and therefore the result obtained is comparable to ones with other methods measurement.

A comparison plot of the Soret coefficient for this mixture can be seen in Figure 3.28. The methods of measurement were the beam deflection method by Kolodner et al. [23] and Zhang et al. [24], TDFRS method used by Kita et al. [27], thermogravitational column and laser Doppler velocimetry by Dutrieux et al. [29] and digital interferometry by Mialdun et al. [31].

The Soret coefficient obtained for the present experiment was $S_T = 4.16 \times 10^{-3} \text{ K}^{-1}$ as mentioned before. As evident from the plot, this value is in between the values obtained by Mialdun and Shevtsova. ($S_T = 3.32 \times 10^{-3} \text{ K}^{-1}$ at $C = 0.491$, $T = 22.5^\circ\text{C}$) and Kita et al. ($S_T = 3.28 \times 10^{-3} \text{ K}^{-1}$ at $C = 0.491$, $T = 22.5^\circ\text{C}$).

Figure 3.28: Plot of various Soret coefficients vs. different water-ethanol mass fractions

$C = 0.5$, $T = 20^\circ\text{C}$), and Dutrieux et al. ($S_T = 5.82 \times 10^{-3} \text{ K}^{-1}$ at $C = 0.5$, $T = 22.5^\circ\text{C}$). The present result gives further confidence in the measurement method and results obtained for other binary mixtures presented in this chapter.

3.3.3. Defining method limitations

The measurement method used does have certain limitations that become evident when mixtures with certain weight percentages are used. Two of these mixtures and their corresponding limitations will be briefly mentioned.

A mixture of presumably water-ethanol 50-50 weight percentage was made, and the experiment was conducted. However, during the data analysis, an abnormal behaviour in the concentration was observed. Therefore, the mixture was checked with a refractometer and it was determined that the mixture was actually water-ethanol 90-10 wt%. It is still interesting to see the behaviour

of this mixture, and a sample of a concentration plot during thermodiffusion is seen in Figure 3.29, where mixing of the liquid is visible, along with convection in the the entire liquid:

Figure 3.29: Water-10wt%ethanol concentration plot.

This behaviour corresponds to a sign change of the Soret coefficient, which leads to instability

Another limitation of the method is evident when the mixture of isopropanol- 5% water is used. The result is presented in the next section; however, it is evident from the data that the method cannot produce good results for this range of weight percentage due to a low dn/dC coefficient resulting in data scatter.

3.3.4. Isopropanol-water mixtures

This section presents the results for the isopropanol-water mixtures with different mass fractions

3.3.4.1 Isopropanol – 50wt% water Mixture

The first mixture used in the experiment was a binary mixture of water 50 wt%-isopropanol. The time evolution plot of the concentration change is presented in Figure 3.30, where the results

coefficients are also stated. In this plot, the fitting curve applied to the experimental data can also be seen.

Figure 3.30: Thermodiffusion plot of concentration change vs. time for water-isopropanol mixture

The resulting Soret coefficient was compared to results from Poty et al. [38] and Mialdun and Shevtsova [28], and plotted as a function of the water mass fraction in Figure 3.31.

As evident from the plot, the current experimental Soret coefficient $S_T = 4.9769 \times 10^{-3} \text{ 1/K}$, for a water mass fraction of 0.5, is between the values obtained by Poty et al., who used the flowing cell method, and Mialdun and Shevtsova who used the current digital interferometry method. It can be safely said that the current experimental Soret coefficient point is acceptable. The experimental result of diffusion coefficient for a water mass fraction of 0.5 was $D = 1.1973 \times 10^{-10} \text{ m}^2/\text{s}$, and is plotted in Figure 3.32. This result is compared to the results obtained by Pratt et al. [39], who used the Taylor dispersion technique, and Mialdun and Shevtsova [28] who used the

Figure 3.31: Soret coefficient of water-isopropanol mixture vs. water mass fraction showing present result, Mialdun et al. [31] and Poty et al. [38]

current digital interferometry method of measurement using a cubic cell. The results of our authors include coefficients obtained at various mass fractions values, and show the general behaviour and trend of the diffusion coefficient as the mass fraction of water is increased. The current experimental result is just below that of Mialdun and Shevtsova. From the plot, it can be seen that this result is at the bottom pool of the results; however, it is evident that it follows the general trend, and is a valid representation of the results for this mixture, at a $\Delta T \approx 5.7\text{K}$.

Figure 3.32: Diffusion coefficient of water-isopropanol mixture as a function of water mass fraction showing present result, Mialdun et al. [28] and Pratt et al. [39]

3.3.4.2 Isopropanol-1 wt% water

The results for the mixture with water mass fraction of 1% are presented in Figure 3.33. The Soret coefficient was determined to be $S_T = 4.90 \times 10^{-4} \text{ 1/K}$, and the diffusion coefficient $D = 2.10 \times 10^{-10} \text{ m}^2/\text{s}$. The resulting coefficients correspond to a layer thickness of only $\Delta z = 2.9 \times 10^{-3} \text{ m}$, due to the instability of the results. Compared to the results of Poty et al. [38], again, the present result does not follow the trend. The results obtained here are considered unreliable, due to a low dn/dC coefficient, and the instability encountered in the results.

Figure 3.33: Thermodiffusion plot of concentration change vs. time for water-1wt% isopropanol mixture

3.3.4.3 Isopropanol-5 wt% water Mixture

From Figure 3.31 it can be seen that the only available data for the Soret coefficient for mixtures in the negative Soret region date back to values obtained in 1974 by Poty et al. [38]. In order to further investigate this region and verify the old results, a mixture of 95 wt% isopropanol-5 wt% water was used (water: 0.9953 g, isopropanol: 19.008 g).

The evolution of concentration change with respect to time is shown in Figure 3.34. As evident, there is a lot of 'noise', or data scatter present, as expected. This region experiences phase separation, and therefore the data are scattered as seen. However, a trend was able to be determined, and the resulting coefficients are $S_T = 6.78 \times 10^{-3} \text{ 1/K}$, $D = 6.94 \times 10^{-10} \text{ m}^2/\text{s}$ for a layer thickness of $\Delta z = 4.1 \times 10^{-3} \text{ m}$. The resulting Soret coefficient is different than the results

expected of one that would follow the trend of the curve from Poty et al. from Figure 3.31, where the result should be expected to be in the negative region. However, the present result is in the positive Soret region. The present diffusion coefficient is in good agreement with previous trends. From Figure 3.32, the present result follows the same trend of the results obtained by Pratt et al. [39]. Since the present method of interferometry is different than the methods used by other investigators, it is necessary to conduct further experiments in the negative Soret region of water-isopropanol mixtures. Although the results for this mixture show better agreement with literature compared to the isopropanol – 1 wt% water mixture, they are still considered unreliable as again this mixture has a low dn/dC coefficient, causing instability in the results.

Figure 3.34: Thermodiffusion plot of concentration change vs. time for isopropanol-5wt% water mixture

3.3.4.4 Isopropanol-15 wt% water Mixture

As the concentration of water is increased and we move away from the unstable region, it can be seen from time evolution plot of concentration difference in Figure 3.35 that the data points

become more stable and are less dispersed, as was evident in the mixtures of 1 and 5 wt% water. The resulting coefficients for this mixture were determined to be $S_T = 2.21 \times 10^{-3} \text{ 1/K}$, $D = 4.43 \times 10^{-10} \text{ m}^2/\text{s}$. These coefficients correspond to a layer thickness of $\Delta z = 3.7 \times 10^{-3} \text{ m}$. Referring to Figures 3.31 and 3.32, it is clearly evident that the results are following the trend as those obtained by Mialdun and Shevtsova [31] for other weight percentages isopropanol-water.

Figure 3.35: Thermodiffusion plot of concentration change vs. time for isopropanol-15wt% water mixture

3.3.4.5 Isopropanol-80 wt% water Mixture

Results could not be produced for this mixture, because the Soret coefficient was found to be in the negative region.

3.3.4.6 Isopropanol-70 wt% water Mixture

At the other end of the plot, moving closer to the negative Soret region once again, it seems from the time evolution plot of the concentration change in Figure 3.36, that the data points are still very stable and make for a nice plot. The resulting coefficients are $S_T = 1.91 \times 10^{-3} \text{ 1/K}$, and $D = 2.32 \times 10^{-10} \text{ m}^2/\text{s}$. For this mixture, the resulting coefficients correspond to a larger layer thickness, $\Delta z = 4.8 \times 10^{-3} \text{ m}$, covering most of the data range of the cell height.

Figure 3.36: Thermodiffusion plot of concentration change vs. time for isopropanol-70wt% water mixture

Overall, the present experimental results are compared to literature results in Figures 3.37 and 3.38, for the Soret and diffusion coefficients respectively:

Figure 3.37: Present results of Soret coefficient of water-isopropanol mixture vs. water mass fraction compared to literature results of Mialdun et al. [31] and Poty et al. [38]

Figure 3.38: Present results of diffusion coefficient of water-isopropanol mixture vs. water mass fraction compared to literature results of Mialdun et al. [31] and Pratt et al. [39]

From the plot of Soret coefficients in Figure 3.37, the present experimental results seem to follow the same trend as the results obtained by Mialdunand Shevtsova, involving the same measurement method. As mentioned previously, the results for the mixtures involving 1wt% and 5wt% of water are considered unreliable due to a low dn/dC coefficient, which causes instability in the results. In addition, as mentioned before, no results were obtained for a mixture containing 80wt% water, due to a negative Soret. When looking at the plot for the experimental Soret coefficients and following the trend, it is evident that the sign change occurs between water mass fractions of 70 and 80wt% of water. The same statement holds true for the diffusion coefficient results, as shown in Figure 3.38.

3.4 Summary

Using a Mach-Zehnder digital optical interferometry, the Soret and diffusion coefficients of the binary mixture isopropanol-water, with different water mass fractions were investigated, in terrestrial conditions. In order to determine the validity and accuracy of this method, it was necessary to perform some experiments for calibrations and verification of the system and of the results. Therefore, binary mixtures with equal mass fractions of water-ethanol, IBB-Dodecane, IBB-THN and THN-Dodecane were used in the experiment. These mixtures have been used in other experiments before, using different and similar measurement methods, and therefore other data and benchmark values from literature are available for comparison. The comparison showed acceptable results, therefore creating confidence in the method. Finally, binary mixtures of isopropanol-water were investigated with varying water mass fractions. These results were then compared to other literature results. Generally, the results showed good agreement with previous trends. The results for water mass fractions of 1 and 5wt% were deemed unreliable, due to low dn/dC coefficients.

In conclusion, it is evident that using digital optical interferometry, it is possible to measure diffusion and Soret coefficients of binary mixtures in the presence of gravitational forces, and obtain good results. In order to further investigate the effect of g-jitter on the binary mixture of water and isopropanol, it was decided to conduct a numerical analysis using a different cell, which was presented in the following section. This experiment is intended to be sent on the International Space Station for further testing as part of the IVIDIL (Influence of Vibrations on Diffusion in binary Liquids) research. The results will then be compared to the numerical and experimental results presented in this thesis.

Chapter 4. Study of the Effects of G-jitter in Liquids

in the Presence of Thermodiffusion

4.1 Introduction

It is necessary to perform the numerical analysis in order to better understand how the flow inside the cell is affected when external vibrations are introduced. Therefore, a comparison is made between the zero-gravity, micro-gravity and g-jitter scenarios. The analysis conducted uses coefficients such as Soret and thermodiffusion coefficients, and g-jitter data from previous experiments to obtain results. The end goal is to be able to make preparations for future IVIDIL (influence of vibrations on diffusion in liquids) experiments, to measure the experimental thermodiffusion coefficient and to compare with the numerical analysis presented in this chapter.

4.2 Model Description

The three dimensional model was created using the commercial software COMSOL. The model is on a xyz-coordinate system and consists of three cylinders. The two end cylinders are 13.5 mm in length, with a diameter of 12 mm. The centre cylinder is 13 mm in length, with a diameter of 6mm. The overall length of the model is 40 mm. A technical drawing of the actual cell with hardware is included in Appendix F. The model created in COMSOL can be seen in Figure 4.1.

The model contains a binary mixture of water-isopropanol, with a 90:10 percentage mass fraction distribution, which is the same mixture and mass fraction that will be experimented on the ISS. It is subjected to a temperature gradient at its end walls. One side of the wall is subjected to an initial temperature of 29.5°C, called T_C . The other side is subjected to an initial temperature

Figure 4.1: Numerical model generated in COMSOL

of 39.5°C , called T_H . The temperature in the centre of the model is set to the average of both T_C and T_H , called T_{avg} .

The temperature gradient to which the model is subjected to will create a temperature and concentration gradient of the mixture. The software COMSOL will incorporate the governing equations of the Navier Stokes, energy and species equations. Several different scenarios are considered in numerical modeling: (1) Two-dimensional model with zero gravity scenario in steady state which can be considered an 'ideal' condition, (2) to an initial temperature of 29.5°C , called T_C . The other side is subjected to an initial temperature Two-dimensional model in a microgravity setting, (3) Three-dimensional model undergoing varying gravitational variations called g-jitter. The g-jitter data is obtained from FOTON-12 TRAMP, and can be regarded as a 'realistic' scenario.

4.3 Mesh Analysis

In the case of the two-dimensional model, several mesh cases were analyzed. In COMSOL, it is stated that for a mesh with triangular elements, a mesh quality ratio of $q > 0.3$ is necessary in order for the quality of the solution not be affected by the quality of the mesh. For a two-dimensional case, the calculations and iterations performed by the solver are less complex. The figure below shows the two-dimensional meshed model with triangular mesh:

Figure 4.2: Two-dimensional model

The mesh statistics for the above meshed model are as shown in Table 4.1:

Table 4.1: Mesh statistics of two-dimensional model

No. of mesh points	No. of elements (triangular)	No. Degrees of freedom	Minimum element quality (q)	Element area ratio
141	236	2209	0.7906	0.2002

The above table shows a mesh with 236 triangular elements and a minimum mesh quality of 0.7906. Other cases with increased mesh were investigated. It was determined that an increased mesh with, for example, 944 elements and the same mesh quality did not result in significant change in the results. Since an increased mesh results in longer computation time and uses more memory, it was decided that the above mesh was sufficient enough for this two dimensional zero-gravity and microgravity cases.

The three-dimensional model presents a more challenging shape to mesh. Similar to the two-dimensional case, several different mesh cases were analyzed. Initial cases involved using the default triangular mesh, starting with low number of elements, and slowly refining the mesh. However, the default triangular mesh was deemed not appropriate for this model, because the results would not converge, especially close to the hot and cold wall edges. Therefore, a custom swept mesh was introduced, as seen in Figure 4.3.

A simple swept mesh was not sufficient enough to get rid of the divergence problems. It was necessary to create a finer mesh near the edges of the model. As visible from the above figure, the vertical walls have triangular meshes, which are then swept in the x-direction, creating prismatic mesh elements. The mesh statistics are in Table 4.2.

Table 4.2: Three-dimensional Mesh statistics

No. of mesh points	No. of elements (prism)	No. Degrees of freedom	Minimum element quality (q)	Element aspect ratio
1140	1780	41415	0.1939	0.0214

Figure 4.3: Three-dimensional model

If the mesh quality $q > 0.1$ for the above mesh with prisms, then the mesh quality does not affect the solution's quality. In this case, as mentioned in the above table, the mesh quality is 0.1939. In addition to this measure, Figure 4.4 depicts the mesh quality throughout the entire model. As evident, the lowest mesh quality is at the far edges of the model, in blue colour, with a minimum q -value of 0.194. Elsewhere in the model, inward from the outer edges, the mesh quality increases to a maximum quality value of 0.849.

Figure 4.4: Three-dimensional mesh quality plot

4.4 Numerical Results

In this section, the numerical results for the three previously mentioned cases are presented and discussed.

4.4.1 Zero gravity case

4.4.1.1 Temperature analysis

The two-dimensional steady state case, as mentioned before, is considered as an ideal case, since there are no residual forces to affect diffusion, and therefore the separation and the results will be perfect. It is advantageous to conduct numerical analysis using this initial ideal model, because it can be used to determine whether the generated model produces the correct results.

Figure 4.5 shows a two-dimensional view of the model at steady state:

Figure 4.5: Two-dimensional temperature and concentration distribution at time 300 hours

The vertical lines visible in the above figure represent the component concentration separation within the cell. It is clearly evident that the lines are perfectly straight, except near the transitions to the narrower cell. This is typical and expected in the ideal case, due to the absence of any other forces that could potentially affect the separation, as will become evident in other cases. In addition to the concentration contour lines, the temperature distribution of the cell is visible. From the left side, the cold wall, the blue shade gradually fades towards the centre. A similar trend is visible from the right side from the hot wall. The scales for both the temperature and concentration are visible on the right side of the figure. Furthermore, it is possible to show when the temperature reaches the steady state in the model, as seen in Figures 4.6 (a)-(c):

(a) Temperature versus time

(b) Temperature versus time

(c) Temperature versus time

Figure 4.6 (a-c): Temperature vs time at points 1:(0.0135,0.006), 2:(0.02,0.006) & 3:(0.0265,0.006) respectively

These figures show the temperature change at three different points throughout the model, all located on the horizontal centre axis, shown in Figure 4.7:

Figure 4.7: Two-dimensional model showing locations of 3 points

As seen from all three figures, the change in temperature reaches a steady state at about a t 3000 seconds (50 minutes). Figure 4.8 shows the temperature along the entire centre axis of model:

Figure 4.8: Temperature across the centre line of the model (x in metres) at four different times

There are three lines in the above plot, each corresponding to a different time 1020, 2010 and 3000 seconds respectively. As evident from this plot, there is not much significant change in temperature along the horizontal axis of the model after 1020 seconds.

4.4.1.2 Concentration analysis

Before the plots for the concentration differences in the model are presented, it is necessary to show the different planes selected in the model to calculate the concentration differences. Figure 4.9 shows the chosen points:

Figure 4.9: 2D model showing 4 point where concentration data was taken

The data was taken at the four points shown in the above figure. Points 1 and 4 are close to the cold and hot wall respectively. Points 2 and 3 are located inline with the smaller portion of the cell. All points are located on the horizontal central axis of the model. Concentration differences will be measured between points 4 and 1 (Point4-Point1), and points 2 and 3 (Point3-Point2).

Figure 4.10 a-b shows the progression of the concentration change in the two-dimensional model, where (b) is a continuation of (a):

(a)

(b)

Figure 4.10(a-b): 2D 0-g concentration differences across the model, and average concentration difference. (b) is a continuation of (a)

In Figure 4.10 (a) & (b), there are three lines shown. As mentioned, the first line, labeled 'Plane4-Plane1' represents the concentration difference between the hot wall and the cold wall. The line labeled 'Plane3-Plane1' represents the concentration difference between the start and end of the narrow part of the cell. The third curve, labeled 'Delta C avg', represents the difference between the averages of the concentration values of points 3 and 4, and points 1 and 2. From Figure 4.10(a), it is evident that the concentration difference continues until it reaches a time of 960000 seconds, equivalent to 266 hours and 40 minutes. Although after this point, there is a small increase in concentration difference still visible, it is very minor. Therefore, it seems that for the zero-gravity, or ideal scenario, more than 266 hours are required in order for the concentration of the mixture to reach a steady state. Furthermore, it should be noted that the change in concentration averages of the hot and cold sides (middle curve) does not surpass a concentration difference of 0.075.

4.4.2 Two-dimensional Micro-gravity case

The two-dimensional micro-gravity case, in the transient state, introduces gravitational force at the micro (10^{-6}) level in equations (2), (3) and (4) for the “ ρg ” term in the y-direction only, where $g = 9.81 \times 10^{-6} \text{ m/s}^2$. The remainder of the conditions and equations remain the same as the two-dimensional zero-gravity case.

The two-dimensional model with microgravity can be seen in Figure 4.11:

Figure 4.11: Two-dimensional microgravity model temperature and concentration distribution at 200 hours

In this case, it is evident from the figure that the contour lines representing the component separation are not as straight anymore. This is a clear indication that the microgravity forces have affected the separation of the two components within the fluid, causing the visible convection.

On the top half of the cell, there is flow from the hot wall to the cold wall, and on the bottom half

of the cell there is a flow from the cold wall to the hot wall. Therefore, there is an overall clockwise flow inside the cell.

The time necessary to reach the steady state for temperature seems also to be affected by the presence of micro gravitational forces. Figure 4.12 shows the progress of temperature at point $(x,y)=(0.0265, 0.006)$ in the model, over a time of 4000 seconds. It is visible from the curve that it takes slightly longer for the curve to reach a steady state when compared to Figure 4.6c, which presented the same temperature progression over time for the same point, without the presence of micro gravitational forces.

Figure 4.12: Temperature versus time at point $(x,y) = (0.0265, 0.006)$

Figure 4.13 shows the temperature distribution along the centre axis of the model at five different times. It is visible that after a time of 2000 seconds, there is not much change in temperature across the model. Although a minor difference is visible in the curves for times 3000 and 3900, this difference in temperature is not large. It does show once again that due to the presence of micro gravitational forces, the temperature distribution remains relatively stable over time.

micro gravitational forces, the time required to reach a steady state for temperature is slightly increased.

Figure 4.13: Temperature across the centre line of the model (x in metres) at different times

4.4.2.1 Velocity contour

Figure 4.14 below shows the velocity contour plot in the x-component of the velocity.

Figure 4.14: Velocity contour plot in x-direction for two-dimensional model

It is evident from Figure 4.14 that there is a symmetrical flow inside the cavity, caused by the presence of micro-gravitational forces.

4.4.3 Three-dimensional G-jitter case

As explained earlier, experiments carried out in space still experience some small residual accelerations termed 'g-jitter'. As mentioned in Chapter 2, the case where g-jitters are introduced into the model is considered the 'real' scenario. The g-jitter data and the corresponding Fourier coefficients are obtained from experiments on board FOTON, and are applied in the x, y and z directions, using equation (6) from Chapter 2, and included in the Navier Stokes equations in COMSOL.

4.4.3.1 Temperature analysis

Figure 4.15a-c below shows the temperature with respect to time at three different points in the model; the same point that were chosen for the two-dimensional model plots. All points are on the horizontal centre axis of the model, with x-values: 0.0135, 0.02 and 0.0265 [m].

(a) Temperature versus time

(b) Temperature versus time

(c) Temperature versus time

Figure 4.15 a-c: Temperature vs time for three different points along the centre x-axis

As evident from the plots, the time necessary for the temperature to reach a steady state is about 6000 seconds. Compared to the two-dimensional zero-gravity case with a time of 3000 seconds,

it can be concluded that with the presence of g-jitter to the model, the time necessary for the temperature to reach a steady state has doubled in value.

4.4.3.2 Concentration analysis

Figure 4.16 shows the concentration distribution of the three-dimensional model:

Figure 4.16: Concentration distribution of three-dimensional g-jitter model at 300 hours

Figure 4.16 represents the concentration distribution across the model at a time of 300 hours. Referring to the colour bar to the right of the image, the lowest level of concentration is represented by the dark blue colour, and the highest level of concentration by a red colour. It is evident from this figure that the heavier component, water, has moved to the hot wall, the right side of the cell. This occurs due to the negative Soret effect.

In order to get a clearer picture of the concentration distribution in the cell, Figure 4.17a-b presents the distribution of concentration with respect to time, where (b) is a continuation of (a).

(a)

(b)

Figure 4.17 a-b: Three-dimensional g-jitter concentration differences across the model, and average concentration difference. (b) is a continuation of (a)

From Figure 4.17a, it is clearly seen that the concentration difference curves are more gradual and take a longer time to reach a steady state. In fact, referring to Figure 4.17b, a continuous increase in concentration difference is seen to the end of the plot, and predictably beyond 300 hours. The difference of the concentration averages of the hot and cold sides seem to be a bit lower, 0.069, than the zero-gravity scenario which was at 0.075, at 300 hours. A comparison of the difference of the concentration averages of the hot and cold sides for the two-dimensional zero-gravity and three-dimensional g-jitter cases can be seen in Figure 4.18:

Figure 4.18: Comparison plot of the difference of concentration averages between the hot and cold sides, for two-dimensional 0-g and three-dimensional g-jitter cases

It can then be stated that the addition of g-jitter has caused an increase in the time required for the concentration change in the mixture to reach a steady state, and that it is necessary to analyze the results beyond 300 hours.

4.4.3.3 Velocity analysis

The velocity profiles for the X-, Y-, and Z-directions are presented, with data corresponding to the centre line of the model at time 294-300 hours. Figure 4.19 a-b shows the velocity plots in the X-direction, where (b) is a zoomed-in view of (a):

Figure 4.19b: Zoomed-in view of X-velocity vs time for three-dimensional g-jitter from time 291.6-300 hours

In Figure 4.19a, the curve represents X-velocity data for the last 8 hours of analysis. As one can see, there are unsteady fluctuations in the velocity values. A closer look at these oscillations can be seen in Figure 4.19b. Figure 4.20 (a) and (b) show the Y-velocity for the last 6 hours and zoomed-in view of the velocity profile respectively.

Figure 4.20a: Y-velocity vs time for three-dimensional g-jitter from time 294-300 hours

Figure 4.20b: Zoomed-in view of Y-velocity vs time for three-dimensional g-jitter from time 294-300 hours

Figure 4.21 (a) represent the velocities in the Z-direction, and (b) is a zoomed-in view of plot (a):

Figure 4.21a: Z-velocity vs time for three-dimensional g-jitter from time 294-300 hours

Figure 4.21b: Zoomed-in view of Z-velocity vs time for three-dimensional g-jitter from time 294-300 hours

It is clearly seen from all velocity plots that although the presence of g-jitter has some effect on velocity, the overall effect can be deemed negligible, due to the small magnitude of the velocities (10^{-9} to 10^{-7}). It can therefore be concluded that the FOTON-M12 platform is suitable to conduct future experimentation. In order to determine the same for the ISS platform, real time data are required from future IVIDIL experiments to determine whether the presence of g-jitter can be deemed negligible as well.

4.5 Summary

Three different scenarios have been analyzed in this chapter: (1) two-dimensional zero-gravity, (2) two-dimensional micro-gravity and (3) three-dimensional g-jitter. The plots for the 'ideal' zero-gravity and 'real' g-jitter scenarios were compared to determine the effect of the presence of g-jitter. In two types of analysis – temperature and concentration – it was concluded that the presence of g-jitter increased the time required for both these parameters to reach a steady state. It was noticed, however, to fully determine the time required for the concentration difference to reach the steady state, it is necessary to conduct numerical analysis beyond 300 hours. In the case of the velocity analysis, the presence of g-jitter introduced oscillations in the velocity profiles. It was concluded, however, that the effects could be regarded as negligible and that the FOTON-M12 platform is suitable for further experiments.

Chapter 5. Conclusion and Future Work

5.1 Summary

Experiments were performed using digital optical interferometry to take measurements of liquid binary mixtures and determine the Soret and diffusion coefficients in terrestrial conditions. The interferometer was based on a Mach-Zehnder setup, which involves the use of several mirrors and lenses which deflect a constant wavelength laser beam. Using a beam splitter, two beams are obtained, one of which travels through the experimental glass cell containing the binary mixture, while the other bypasses the cell, uninterrupted. The two beams are then reunited and captured by a camera, connected to a computer which records the images, which show dark and light lines called fringes. From these images, using code written in MATLAB, the change in concentration within the liquid is determined, and the resulting Soret and diffusion coefficients are calculated. The method of measurement was validated using several binary mixtures and comparing the results with previously published benchmark data.

Furthermore, a numerical analysis was performed on a binary liquid mixture of water-isopropanol using the commercial finite element software COMSOL. A three-dimensional cell was created, and the microgravity environment was simulated, with the presence of g-jitter, using real g-jitter data from FOTON. The effect of the presence of g-jitter in the x, y and z-directions on the mixture was studied and compared to a two-dimensional model in a zero-gravity scenario. Numerical results showed that the presence of g-jitter affected the time required for the temperature and concentration change in the mixture to reach a steady state. In addition, fluctuations in velocity profiles, more noticeable in the y-direction, were noticed.

In conclusion, it can be stated with confidence that digital optical interferometry is a competitive and accurate method to measure and determine the transport coefficients of liquid binary mixtures. The method of measurement was validated by measuring the Soret and diffusion coefficients of several liquid binary mixtures and comparing with published results, with good agreement. Then, measurements of the binary mixture of water-isopropanol in a rectangular glass cell were taken at different weight fractions. The Soret and diffusion coefficients were compared to published results. The present results showed good overall agreement and followed the same trend as other sets of data.

Furthermore, the commercial finite element software COMSOL can be used successfully to perform a numerical analysis to determine the effects of g-jitter on a liquid binary mixture of water-10% isopropanol in microgravity conditions. The presence of g-jitter caused increased times in temperature and concentrations to reach a steady state. In addition, it caused fluctuations in the velocity profiles, most noticeable in the y-direction.

5.3 Future Work

- Perform further three-dimensional numerical analysis with the same binary mixture, this time with varying diffusion coefficients, and compare to future experiments conducted in the International Space Station using IVIDIL apparatus.
- Conduct experiments using digital optical interferometry using two different light sources with different wavelengths and ternary mixture, because determining the transport coefficients of ternary mixtures will better represent the fluid behaviour of hydrocarbons in oil reservoirs.

References

- [1] H. Li, L. Wu, On the relationship between thermal diffusion and molecular interaction energy in binary mixtures, *J. Phys. Chem. B*, 108, 2004, p. 13821
- [2] E.W. Haddon, N. Riley, The heat transfer between concentric vibrating circular cylinders, *Quart. J. Mech. Appl. Math* 34, 1981, p. 345
- [3] B.J. Davidson, Heat transfer from a vibrating circular cylinder, *Internat. J. Heat Mass Transfer* 16, 1973, p. 1703
- [4] J.H. Merkin, Oscillatory free convection from an infinite horizontal cylinder, *J. Fluid Mech.* 30, 1967, p. 561
- [5] M. Chacha, D. Faruque, M.Z. Saghir, J.C. Legros, Solutal thermodiffusion in binary mixture in the presence of g-jitter, *Internat. J. Thermal Sciences* 41, 2002, p. 899
- [6] R. Savino, Residual-g and g-jitter effects on the measurement of thermophysical properties in microgravity, *Adv. Space Res.* 29 (4), 2002, p. 559
- [7] Y. Yan, V. Shevtsova, M.Z. Saghir, Numerical study of low frequency g-jitter effect on thermal diffusion, *Fluid Dynamics and Mat. Processing* 4 (1), 2005, p. 315
- [8] Y. Yan, K. Jules, M.Z. Saghir, A comparative study of g-jitter on thermal diffusion aboard the International Space Station, *Fluid Dynamics and Mat. Processing*
- [9] Y. Yan, S. Pan, K. Jules, M.Z. Saghir, Vibrational effect on thermal diffusion under different microgravity environments, *Microgravity Science and Technology* 19 (2), 2005, p. 12
- [10] M.. Chacha, M.Z. Saghir, Solutal-thermo-diffusion convection in a vibrating rectangular cavity, *Internat. J. Thermal Sciences* 44, 2005, p. 1
- [11] M. Chacha, M.Z. Saghir, A. Viviani, 3D numerical simulations of thermodiffusion experiment for a ternary mixture on-board Foton, *Microgravity Science and Technology* 17 (2), p. 31

- [12] V. Shevtsova, D. Melnikov, J.C. Legros, Y. Yan, M.Z. Saghir, T. Lyubimova, G. Sedelnikov, B. Roux, Influence of vibrations on thermodiffusion in binary mixture: a benchmark of numerical solutions, *Physics of Fluids* 19, 2007, 017111
- [13] V. Shevtsova, D. Melnikov, A. Mialdun, J. C. Legros, Development of convection in mixture with Soret effect, *Microgravity Science and Technology* 18 (3-4), 2006, p. 38
- [14] M. Hipp, J. Woisetschläger, P. Reiterer, T. Neger, Digital evaluation of interferograms, *Measurement* 36, 2004, p.53
- [15] Z. Liu, M. Centurion, G. Panotopoulos, J. Hong, D. Psaltis, Holographic recording of fast events on a CCD camera, *Optic Letters*, 27(1), 2002, p.22
- [16] C.R. Coggrave, Quantitative Interferogram Analysis, <http://www.phasevision.com/>
- [17] W.B. Li, P.N. Segre, R.W. Gammon, J.V. Sengers, M. Lamvik, Determination of the temperature and concentration dependence of the refractive index of a liquid mixture, *Journal of Chemical Physics* 101(6), 1994, p. 5058
- [18] G.S. Spagnolo, D. Paoletti, D. Ambrosini, V. Bagini, M. Santarsiero, Fourier transform evaluation of digital interferograms for diffusivity measurement, *Pure Appl. Opt.* 3, 1994, p. 249
- [19] N. Rashidnia, R. Balasubramaniam, Development of an interferometer for measurement of the diffusion coefficient of miscible liquids, *Applied Optics* 41(7), 2002, p. 1337
- [20] P. Kolodner, H. Williams, C. Moe, Optical measurement of the Soret coefficient of ethanol/water solutions, *Journal of Chemical Physics* 88(10), 1988, p. 6512
- [21] K.J. Zhang, M.E. Briggs, R.W. Gammon, J.V. Sengers, Optical measurement of the Soret coefficient and the diffusion coefficient of liquid mixtures, *Journal of Chemical Physics* 104(17), 1996, p. 6881
- [22] W. Köhler, B. Müller, Soret and mass diffusion coefficients of toluene/n-hexane mixtures, *J. Chem. Phys.* 103(10), 1995, p. 4367

- [23] O. Ecenarro, J.A. Madariaga, J. Navarro, C.M. Santamaria, J.A. Carrion, J.M. Saviron, Fickian and thermal diffusion coefficients from liquid thermogravitational columns, *J. Phys. Condens. Matter* 2, 1990, p. 2289
- [24] R. Kita, S. Wiegand, J. Luettmner-Strathmann, Sign change of the Soret coefficient of poly(ethylene oxide) in water/ethanol mixtures observed by thermal diffusion forced Rayleigh scattering, *Journal of Chemical Physics* 23(8), 2004, p.3874
- [25] S. Wiegand, W. Köhler, Measurement of transport coefficients by an optical grating technique, *LNP* 584, 2002, p. 189
- [26] J.F. Dutrieux, J.K. Platten, G. Chavepeyer, M.M. Bou-Ali, On the measurement of positive Soret coefficients, *J. Phys. Chem.* 106, 2002, p. 6104
- [27] G.S. Spagnolo, D. Ambrosini, D. Paoletti, Liquid diffusion coefficients by digital moiré, *Opt. Eng.* 43(4), 2004, p. 798
- [28] A. Mialdun, V. Shevtsova, Development of optical digital interferometry technique for measurement of thermodiffusion coefficients, *Int. J. Heat Mass Transfer* 51 (11-12), 2008, p. 3164
- [29] J.K.Platten, M.M. Bou-Ali, P. Costesèque, J.F. Dutrieux, W. Köhler, C. Leppla, S. Wiegand, G. Wittko, Benchmark values for the Soret, thermal diffusion and diffusion coefficients of three binary organic liquid mixtures, *Philosophical Magazine* 83(17-18), 2003, p. 1965
- [30] G. Wittko, W. Köhler, Precise determination of the Soret, thermal diffusion and mass diffusion coefficients of binary mixtures of dodecane, isobutylbenzene and 1,2,3,4-tetrahydronaphthalene by a holographic grating technique, *Philosophical Magazine* 83(17-18), 2003, p. 1973
- [31] F.J. Bonner, L.O. Sundelof, Thermal diffusion as a mechanism for biological transport, *Z. Naturforsch. C* 39, 1984, p. 656
- [32] <http://www.ucalgary.ca/~schramm/mirror.htm>

- [33] J.K. Platten, The Soret effect: A review of recent experimental results, *Journal of Applied Mechanics* 73, 2006, p. 5
- [34] Cooley, James W., and John W. Tukey, "An algorithm for the machine calculation of complex Fourier series," *Math. Comput.* 19, 297–301 (1965).
- [35] B.R. Hammond, R.H. Stokes, Diffusion in binary liquid mixtures. Part 1. Diffusion coefficients in the system ethanol+water at 25, *J. Chem. Soc., Faraday Trans.* 49, 1953, p. 890
- [36] K.R. Harris, T. Goscinska, H.N. Lam, Mutual diffusion coefficients for the systems water-ethanol and water-propan-1-ol at 25°C, *J. Chem. Soc., Faraday Trans.* 89(12), 1993, p. 1969
- [37] K.C. Pratt, W.A. Wakeham, The mutual diffusion coefficients of ethanol-water mixtures: determination by rapid, new method, *Proc. R. Soc. Lond. Ser. A – Math. Phys. Sci.* 336, 1974, p. 393
- [38] P. Poty, J.C. Legros, G. Thomaes, Thermal diffusion in some binary liquid mixtures by the flowing cell methods, *Z. Naturforsch.* 29A, 1974, p. 1915
- [39] K.C. Pratt, W.A. Wakeham, The mutual diffusion coefficient for binary mixtures of water and the isomers of propanol, *Proc. R. Soc. Lond. Ser. A – Math. Phys. Sci.* 342, 1975, p. 401

Appendices

Appendix A: COMSOL 3D G-jitter Model Input File

3D Gjitter

A.1. Table of Contents

- Title - 3D Gjitter
- Table of Contents
- Model Properties
- Constants
- Geometry
- Geom2
- Solver Settings
- Postprocessing
- Variables

A.2. Model Properties

Property	Value
Model name	3D Gjitter
Author	Pouyan Ezzatian
Company	Ryerson University
Department	Mechanical Engineering

Reference	
URL	
Saved date	Aug 28, 2008 4:07:52 PM
Creation date	Jan 21, 2008 1:55:31 PM
COMSOL version	COMSOL 3.4.0.248

File name: /home/grad/pezzatian/Comsol Analysis/Models/3D g-jitter with rho/3D Gjitter Models from 690-1080/1050-1080k s.mph

Application modes and modules used in this model:

- **Geom2 (3D)**
 - Convection and Conduction
 - Incompressible Navier-Stokes
 - PDE, General Form

A.2.1. Model description

3D model with g-jitter in all directions, with g-jitter data from FOTON.

A.3. Constants

Name	Expression	Value	Description
Th	39.5[degC]		Hot wall [deg.K]
Tc	29.5[degC]		Cold wall [deg.K]
rho_init	984.00[kg/m^3]		Density [kg/m3]
Dc	8.7e-10[m^2/s]		Mass Diff [m2/s]
Dt	St*Dc		Therm Diff [m2/s K]
Pinitial	101000[Pa]		Presssure [Pa]
Tavg	(Th+Tc)/2		Average Temp [deg.K]
St	-10.6e-3[1/K]		Soret coeff.

Beta_T	3.1e-4[1/K]		Thermal expansion coeff
Beta_c	0.1386		Volumetric conc exp coeff
a0x	-3.6615e-6[m/s^2]		Fourier coeff. in x-direction
a1x	-24.896e-6[m/s^2]		Fourier coeff. in x-direction
b1x	30.786e-6[m/s^2]		Fourier coeff. in x-direction
f1x	1.645209e-3[Hz]		Frequency
a0y	-116.65e-6[m/s^2]		Fourier coeff. in y-direction
a1y	1.5392e-6[m/s^2]		Fourier coeff. in y-direction
a2y	3.3976e-6[m/s^2]		Fourier coeff. in y-direction
a3y	4.1744e-6[m/s^2]		Fourier coeff. in y-direction
a4y	-4.7073e-6[m/s^2]		Fourier coeff. in y-direction
a5y	-.96563e-6[m/s^2]		Fourier coeff. in y-direction
b1y	0.42332e-6[m/s^2]		Fourier coeff. in y-direction
b2y	-7.5033e-6[m/s^2]		Fourier coeff. in y-direction
b3y	1.4193e-6[m/s^2]		Fourier coeff. in y-direction
b4y	-2.6550e-6[m/s^2]		Fourier coeff. in y-direction
b5y	-1.4247e-6[m/s^2]		Fourier coeff. in y-direction
f1y	1.157556e-3[Hz]		Frequency
f2y	2.51535e-3[Hz]		Frequency
f3y	1.872925e-3[Hz]		Frequency
f4y	2.237846e-3[Hz]		Frequency
f5y	3.281500e-3[Hz]		Frequency

a0z	16.508e-6[m/s^2]		Fourier coeff. in z-direction
a1z	5.5583e-6[m/s^2]		Fourier coeff. in z-direction
a2z	-0.45562e-6[m/s^2]		Fourier coeff. in z-direction
a3z	-1.5791e-6[m/s^2]		Fourier coeff. in z-direction
a4z	-2.8356e-6[m/s^2]		Fourier coeff. in z-direction
a5z	-0.91597e-6[m/s^2]		Fourier coeff. in z-direction
b1z	2.8422e-6[m/s^2]		Fourier coeff. in z-direction
b2z	-0.70390e-6[m/s^2]		Fourier coeff. in z-direction
b3z	-0.094072e-6[m/s^2]		Fourier coeff. in z-direction
b4z	1.0229e-6[m/s^2]		Fourier coeff. in z-direction
b5z	-0.27967e-6[m/s^2]		Fourier coeff. in z-direction
f1z	1.643534e-3[Hz]		Frequency
f2z	1.870828e-3[Hz]		Frequency
f3z	2.073682e-3[Hz]		Frequency
f4z	3.290399e-3[Hz]		Frequency
f5z	4.149568e-3[Hz]		Frequency

A.4. Geometry

Number of geometries: 1

A.4.1. Geom2

A.4.1.1. Point mode

A.4.1.2. Edge mode

A.4.1.3. Boundary mode

A.4.1.4. Subdomain mode

A.5. Geom2

Space dimensions: 3D

Independent variables: x, y, z

A.5.1. Expressions

A.5.1.1. Subdomain Expressions

Subdomain		1-5
rho		$\text{rho_init} \cdot (1 - \text{Beta_T} \cdot (T - T_{\text{avg}}) + \text{Beta_c} \cdot (-0.9 + c))$
gx	m/s^2	$a0x + a1x \cdot \cos(2 \cdot \pi \cdot f1x \cdot t) + b1x \cdot \sin(2 \cdot \pi \cdot f1x \cdot t)$
gy	m/s^2	$a0y + a1y \cdot \cos(2 \cdot \pi \cdot f1y \cdot t) + b1y \cdot \sin(2 \cdot \pi \cdot f1y \cdot t) + a2y \cdot \cos(2 \cdot \pi \cdot f2y \cdot t) + \dots$ $\dots b2y \cdot \sin(2 \cdot \pi \cdot f2y \cdot t) + a3y \cdot \cos(2 \cdot \pi \cdot f3y \cdot t) + b3y \cdot \sin(2 \cdot \pi \cdot f3y \cdot t) + \dots$ $\dots a4y \cdot \cos(2 \cdot \pi \cdot f4y \cdot t) + b4y \cdot \sin(2 \cdot \pi \cdot f4y \cdot t) + a5y \cdot \cos(2 \cdot \pi \cdot f5y \cdot t) + \dots$ $\dots b5y \cdot \sin(2 \cdot \pi \cdot f5y \cdot t)$
gz	m/s^2	$a0z + a1z \cdot \cos(2 \cdot \pi \cdot f1z \cdot t) + b1z \cdot \sin(2 \cdot \pi \cdot f1z \cdot t) + a2z \cdot \cos(2 \cdot \pi \cdot f2z \cdot t) + \dots$ $\dots b2z \cdot \sin(2 \cdot \pi \cdot f2z \cdot t) + a3z \cdot \cos(2 \cdot \pi \cdot f3z \cdot t) + b3z \cdot \sin(2 \cdot \pi \cdot f3z \cdot t) + \dots$

		$\dots a_4 z \cos(2\pi f_4 z t) + b_4 z \sin(2\pi f_4 z t) + a_5 z \cos(2\pi f_5 z t) + \dots$ $\dots b_5 z \sin(2\pi f_5 z t)$
--	--	---

A.5.2. Mesh

A.5.2.1. Mesh Statistics

Number of degrees of freedom	41415
Number of mesh points	1140
Number of elements	1780
Tetrahedral	0
Prism	1780
Hexahedral	0
Number of boundary elements	868
Triangular	328
Quadrilateral	540
Number of edge elements	276
Number of vertex elements	32
Minimum element quality	0.194
Element volume ratio	0.021

A.5.3. Application Mode: Convection and Conduction (cc)

Application mode type: Convection and Conduction

Application mode name: cc

A.5.3.1. Application Mode Properties

Property	Value
Default element type	Lagrange - Quadratic
Analysis type	Transient
Frame	Frame (xyz)
Weak constraints	Off
Constraint type	Ideal

A.5.3.2. Variables

Dependent variables: T

Shape functions: `shlag(2,'T')`

Interior boundaries not active

A.5.3.3. Boundary Settings

Boundary		2-3, 7, 10-11, 13-19, 23, 26	1, 4	27-28
Type		Thermal insulation	Temperature	Temperature
name		cell	cold wall	hot wall
Temperature (T0)	K	0	Tc	Th

A.5.3.4. Subdomain Settings

Subdomain		1-5
name		cell
Thermal conductivity (k)	W/(m · K)	0.522
Density (rho)	kg/m ³	rho
Heat capacity at constant pressure (C)	J/(kg · K)	3990.50
x-velocity (u)	m/s	u
y-velocity (v)	m/s	v
z-velocity (w)	m/s	w

Subdomain initial value		1-5
Temperature (T)	K	Tc+(Tavg-Tc)*flc2hs(x-0.001,0.002)+(Th-Tavg)*flc2hs(x-0.039,0.002)

A.5.4. Application Mode: Incompressible Navier-Stokes (ns)

Application mode type: Incompressible Navier-Stokes

Application mode name: ns

A.5.4.1. Application Mode Properties

Property	Value
Default element type	Lagrange - P ₂ P ₁
Analysis type	Transient
Corner smoothing	Off
Frame	Frame (xyz)
Weak constraints	Off
Constraint type	Ideal

A.5.4.2. Variables

Dependent variables: u, v, w, p, nxw, nyw, nzw

Shape functions: shlag(2,'u'), shlag(2,'v'), shlag(2,'w'), shlag(1,'p')

Interior boundaries not active

A.5.4.3. Point Settings

Point	1-5, 7-32	6
pnton	0	1

A.5.4.4. Boundary Settings

Boundary	1-4, 7, 10-11, 13-19, 23, 26-28
Type	Wall
name	cell

A.5.4.5. Subdomain Settings

Subdomain		1-5
Integration order (gporder)		4 4 4 2
Constraint order (cporder)		2 2 2 1
name		cell
Density (rho)	kg/m ³	rho
Dynamic viscosity (eta)	Pa · s	.00138744
Volume force, x-dir. (F_x)	N/m ³	gx*rho
Volume force, y-dir. (F_y)	N/m ³	gy*rho
Volume force, z-dir. (F_z)	N/m ³	gz*rho

A.5.5. Application Mode: PDE, General Form (g)

Application mode type: PDE, General Form

Application mode name: g

A.5.5.1. Application Mode Properties

Property	Value
Default element type	Lagrange - Quadratic
Wave extension	Off
Frame	Frame (xyz)
Weak constraints	Off

A.5.5.2. Variables

Dependent variables: c, c_t

Shape functions: shlag(2,'c')

Interior boundaries not active

A.5.5.3. Boundary Settings

Boundary	1-4, 7, 10-11, 13-19, 23, 26-28
Type	Neumann boundary condition
name	cell

A.5.5.4. Subdomain Settings

Subdomain	1-5
name	cell
Source term (f)	$-u*cx-v*cy-w*cz$
Conservative flux source term (ga)	$\{-Dc*cx-Dt*T_x;-Dc*cy-Dt*T_y;-Dc*cz-Dt*T_z\}$
Subdomain initial value	1-5
c	0.9

A.6. Solver Settings

Solve using a script: off

Analysis type	Transient
Auto select solver	On
Solver	Time dependent
Solution form	Automatic
Symmetric	auto
Adaption	Off

A.6.1. Direct (UMFPACK)

Solver type: Linear system solver

Parameter	Value
Pivot threshold	0.1
Memory allocation factor	0.7

A.6.2. Time Stepping

Parameter	Value
Times	1050000:30:1080000
Relative tolerance	0.01
Absolute tolerance	0.0010
Times to store in output	Specified times
Time steps taken by solver	Free
Manual tuning of step size	Off
Initial time step	1e-9
Maximum time step	1.0
Maximum BDF order	5
Singular mass matrix	Maybe
Consistent initialization of DAE systems	Backward Euler
Error estimation strategy	Include algebraic
Allow complex numbers	Off

A.6.3. Advanced

Parameter	Value
Constraint handling method	Elimination
Null-space function	Automatic
Assembly block size	5000
Use Hermitian transpose of constraint matrix and in symmetry detection	Off
Use complex functions with real input	Off
Stop if error due to undefined operation	On
Store solution on file	Off
Type of scaling	Automatic
Manual scaling	
Row equilibration	On
Manual control of reassembly	Off
Load constant	On
Constraint constant	On
Mass constant	On
Damping (mass) constant	On
Jacobian constant	On
Constraint Jacobian constant	On

A.7. Postprocessing

Appendix B: Cell technical drawing (Experimental)

Figure B.1: Technical drawing of the experimental glass cell [Universite Libre Bruxelles – MRC]

Appendix C: Chemical properties

1,2,3,4-Tetrahydronaphthalene

Synonyms: THN; Tetrahydronaphthalene; Tetraline

Molecular Formula: C₁₀H₁₂

Registry number: 119-64-2

MDL number: MFCD00001733

Density: 0.973

Melting point: -35 °C

Risk Description

R19 May form explosive peroxides.

R36/38 Irritating to eyes and skin.

Safety Description

S26 In case of contact with eyes, rinse immediately with plenty of water and seek medical advice.

S28A After contact with skin, wash immediately with plenty of water.

IR

Analysis

Result

433 (49.65); 742 (4.61); 804 (57.18); 944 (60.7); 1111

Miscellaneous information

EINECS 204-340-2

Origin synthetic

Solubility in water:

Product 14673
1,2,3,4-Tetrahydronaphthalene, 98+%

General Product Data

Version	4
CAS No	119-64-2
Molecular weight	132.20

Product Specifications

Appearance	Clear colorless liquid which darkens on storage
Infrared spectrometry	Authentic
Separat. techn. GC	>98.0 %

Isobutylbenzene

Synonyms: 2-Methyl-1-phenylpropane

Molecular Formula: C₁₀H₁₄

Registry number: 538-93-2

MDL number: MFCD00008936

Density: 0.85

Melting point: -51 °C

Risk Description

R10 Flammable.

Safety Description

S16 Keep away from sources of ignition - No smoking.

Miscellaneous information

EINECS 208-706-2

Origin synthetic

Solubility in water:
INSOLUBLE IN WATER

ACROS
organics

Product Specifications

Product 15096
Isobutylbenzene,99.50%

General Product Data

Version 2

CAS No 538-93-2

Molecular weight 134.22

Molecular
formula C₁₀ H₁₄

Linear formula C₆H₅CH₂CH(CH₃)₂

Product Specifications

Appearance CLEAR COLOURLESS LIQUID

Infrared spectrometry AUTHENTIC

Dodecane

Synonyms: n-Dodecane

Molecular Formula: C₁₂H₂₆

Registry number: 112-40-3

MDL number: MFCD00008969

Density: 0.753

Melting point: -12 °C

Risk Description	
R65	Harmful: may cause lung damage if swallowed.
Safety Description	
S36	Wear suitable protective clothing.
S60	This material and its container must be disposed of as hazardous waste.
IR	
Analysis	Result
Miscellaneous information	
EINECS	203-967-9
Origin	synthetic
	Solubility in water:

Product Specifications

Product 11759
Dodecane,99%

General Product Data

Version	5
CAS No	112-40-3
Molecular weight	170.34
Molecular formula	C12 H26
Linear formula	CH3(CH2)10CH3

Product Specifications

Appearance	Clear liquid
Color scale	<10 APHA
Infrared spectrometry	Authentic

P/7500/15	1 L	GB
P/7500/17	2.5 L	GB
P/7500/PB17	2.5 L	PCG
P/7500/PC17	2.5 L	PL
P/7500/21	5 L	MC
P/7500/25†	25 L	MD

† See page 507 for suitable tap

 $(\text{CH}_3)_2\text{CHOH}$

M.W. 80.10

Physical constants

Boiling point	82.5°C
Refractive index	1.3772
Weight per mL at 20°C	0.786 g

Guaranteed analysis

Assay (GLC)	>99.8%
Acidity/Alkalinity	<0.0001 meq/g
C ₄ alcohols	<0.005%
C ₅ alcohols	<0.005%
Colour	<5 APHA
Residue after evaporation	<0.001%
Substances darkened by H ₂ SO ₄	<10 APHA
Substances reducing KMnO ₄	<5 ppm
Water (Karl Fischer)	<0.1%
Calcium (Ca)	<0.2 ppm
Copper (Cu)	<0.02 ppm
Iron (Fe)	<0.1 ppm
Lead (Pb)	<0.02 ppm
Magnesium (Mg)	<0.1 ppm
Potassium (K)	<0.2 ppm
Sodium (Na)	<1 ppm
Zinc (Zn)	<0.1 ppm
Total phosphorus (P)	<0.1 ppm
Total silicon (Si)	<0.05 ppm
Total sulfur (S)	<0.5 ppm
Ethanol	<0.005%
Methanol	<0.005%
Propan-1-ol	<0.05%

Typical analysis

Aluminium (Al)	<0.02 ppm
Barium (Ba)	<0.02 ppm
Cadmium (Cd)	<0.02 ppm
Chromium (Cr)	<0.02 ppm
Cobalt (Co)	<0.02 ppm
Lithium (Li)	<0.02 ppm
Manganese (Mn)	<0.02 ppm
Molybdenum (Mo)	<0.02 ppm
Nickel (Ni)	<0.02 ppm
Strontium (Sr)	<0.02 ppm
Titanium (Ti)	<0.02 ppm
Vanadium (V)	<0.02 ppm

Appendix D: MATLAB Code

The below program is an example of the code for one of the files used to analyze the results using MATLAB 7.1. This particular code converts the raw images obtained from the experiment into images showing the concentration change over time.

```
% Written by A. Mialdun
% Modified by P. Ezzatian

clear all,

% loading the properties data
L = 18e-3; % optical path in liquid [m]
lambda = 632.8e-9; % wavelength [m]
T_top = 28.0; % temperature of the top plate [oC]
T_bot = 22.0; % temperature of the bottom plate [oC]
T_0 = (T_top+T_bot)/2;
C_0 = 5;
%[dn_dC,dn_dT] = refr_ind_eth(C_0,T_0);
%dn_dT = -3.9e-4; % isopropanol-5.0wt.%water (Wiegand)
dn_dC = 0.44e-4; % isopropanol-5.0wt.%water (Wiegand) 4.06

% values to be pointed out manually
ImPath = 'C:\Thesis_2008\Experiments\20080626\images\'; % path to images
Fref = [ImPath 'ima046.bmp']; % image for reference phase extraction
N_ima = [46:243]; % number of image(s) to be processed 122:304

% Selecting image regions for processing
% 1028pxls=18mm, 360pxls=6.3mm, 382pxls=6.7mm
rect_pha = [172 100 1023 511];
rect_puw = [1 58 1024 342]; % for C
if exist('rect_puw','var')==0,
    rect_puw = select_ROI(Fref, rect_pha); % selecting region for unwrapping
end

% reference image processing
I = image_prep(Fref, rect_pha);
[refPha, maxX, maxY] = interf2phase(I);

for i = 1:length(N_ima);
    % name of the file to be processed :
    Fprc = [ImPath 'ima' sprintf('%3d',N_ima(i)), ' ', '0') '.bmp'];

    I = image_prep(Fprc, rect_pha);
    Pha = interf2phase(I, maxX, maxY); % image processing
    Pha = Pha - refPha; % reference phase subtraction
    Pha(Pha < 0) = Pha(Pha < 0) + 2*pi; % ranging to modulo 2*pi

    %_Selecting region for unwrapping
    Pha = Pha(rect_puw(2):rect_puw(2)+rect_puw(4)-1,
rect_puw(1):rect_puw(1)+rect_puw(3)-1);

    % Phase unwrapping
    Pha = puw_gpm(Pha);
    Pha = filt_median(Pha);
```

```

% trimming of X% overshoot pixels
Pha = trimover(Pha, 0.1);

% conversion to refractive index variation
Dn = Pha*lambda/(2*pi*L); % in liquid

% conversion to concentration
DC = -Dn/dn_dC;
DC = DC - mean(DC(:)) + C_0;

dC = mean(DC(:,505:515));
fid = fopen(['C_' strcat(sprintf('%3d',N_ima(i)), ' ', '0') '.txt'],'w');
% overwrite 'w' or add 'a'
fprintf(fid, '%1.4f\n', dC);
fclose(fid);

plot_2D_C_save,
end

%fid = fopen('dC_t.txt','a'); % overwrite 'w' or add 'a'
%fprintf(fid, '%3d %1.4f\n', [N_ima; dC_t]);
%fclose(fid);

```


Figure E.1: Technical drawing of the cell used in numerical analysis. [Verhaert Space]