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### INFLUENCE OF VIBRATION ON DIFFUSION IN LIQUID MIXTURES ON-BOARD THE INTERNATIONAL SPACE STATION

By

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A dissertation presented to

### **Ryerson University**

in partial fulfillment of the

requirements for the degree of

### **DOCTOR OF PHILOSOPHY**

in the Program of

### **MECHANICAL ENGINEERING**

Toronto, Ontario, Canada, 2013

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## Abstract

# Influence of Vibration on Diffusion in Liquid Mixtures On-Board the International Space Station Doctor of Philosophy ARAM PARSA Mechanical Engineering Ryerson University 2013

Experiments on-board the International Space Station experience a convective flow due to the oscillatory g-jitters induced by several sources such as crew activities, mechanical systems, thrusters firing, spacecraft docking, etc. Although g-jitter seems to have a major impact on diffusion-related experiments in Space, very few experimental studies have addressed this topic.

This study examined the effect of oscillatory g-jitters on transport processes (fluid flow, heat transfer and mass transfer). Cubic rigid cells filled with water and isopropanol at different concentrations were subjected to thermal gradients and forced vibrations. The cells were exposed to different levels of vibration in terms of frequency and amplitude, which were applied perpendicular to the temperature gradient. The full transient Navier Stokes equations coupled with the mass and heat transfer formulas were solved numerically using the control volume technique. The physical properties of the fluid mixture such as the density were determined using two different models.

The effect of different levels of vibration on the flow was analysed and the results were compared in a benchmark study with other scientific groups. The effect of the diffusion coefficients variation and other physical properties on the temperature and concentration distribution was compared to those results obtained with constant diffusion coefficients. Results show that use of variable physical properties in the modelling produces different flow patterns and component concentration. By examining different flow patterns, it was found that the effect of using variable coefficients is much more significant in the cases with high Rayleigh vibration

that result in strong flow when compared with numerical analysis using constant variables. The numerical analysis was also performed for the actual experiment on board the International Space Station. The same trend was seen for both the numerical and experimental results. However, the separation of components was higher in the experiment in comparison with the numerical analysis. This was discussed in detail for various scenarios in terms of the applied frequency and amplitude. Recommendations are made according to the findings from this study for the improvement of accuracy in the numerical analyses of future diffusion experiments in Space.

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# Nomenclature

$C_0$	initial water concentration (carrier)	
С	transported component mass fraction	
CP	mixture specific heat	$J \cdot kg^{-1} \cdot K^{-1}$
$D_{C}$	mass diffusion coefficient	$m^2 \cdot s^{-1}$
$D_M$	molecular diffusion coefficient	$m^2 \cdot s^{-1}$
$D_{T}$	thermal diffusion coefficient	$m^2 \cdot s^{-1} \cdot K^{-1}$
f	frequency of the g-jitter	Hz
ω	Angular frequency	s <sup>-1</sup>
$g_{\rm st}$	static residual gravity	$\mathbf{m} \cdot \mathbf{s}^{-2}$
$g_{ m vib}$	amplitude of the oscillatory g-jitter	m
А	amplitude of controlled vibration	m
Ra	Rayleigh vibration number	[1]
$\beta_{\rm C}$	solutal volume expansion coefficient	[1]
$\beta_{T}$	thermal volume expansion coefficient	$\mathbf{K}^{-1}$
J	diffusion flux	kg· m <sup>-2</sup> · s <sup>-1</sup>
q	Heat Flux	W
k	mixture thermal conductivity	$W \cdot m^{-1} \cdot K^{-1}$
L	cavity length	mm
Р	pressure	Pa
$\mathbf{S}_{\mathrm{T}}$	Soret coefficient	$\mathbf{K}^{-1}$
t	time	S
$t_{vs}$	viscous time	S
t <sub>th</sub>	thermal time	S
$t_D$	diffusion time	S
T	temperature	K
$T_0$	room/initial temperature	K
T <sub>C</sub>	cold wall temperature	K
$T_{\rm H}$	hot wall temperature	K
I <sub>m</sub>	Average temperature	K _1
u	velocity component in x-direction	m· s -1
v	velocity component in y-direction	$\mathbf{m} \cdot \mathbf{s}^{-1}$
μ	mixture dynamic viscosity	$kg \cdot m^{-1} \cdot s^{-1}$
ν	mixture kinematic viscosity	$m^2 \cdot s$
ρ	mixture mass density	kg· m <sup>-3</sup>
χ	thermal diffusivity	$m^2 \cdot s$

# Subscripts:

0	reference state or initial state
cold	at the cold wall
hot	at the hot wall
VS	viscous
th	thermal

# **Chapter 1**

## Introduction

Molecular diffusion takes place in a mixture due to concentration gradients. Separation of the constituents in a mixture is enhanced by temperature gradients through the process of thermal diffusion. Diffusion is the result of the combined effects of temperature and concentration. Therefore, the Soret coefficient, defined as  $S_T = D_T/D_M$ , measures the separation of components in a mixture under the influence of a temperature difference. Steady state can be obtained when the separating effect of thermal diffusion is balanced by the remixing effect of molecular diffusion. Thermal diffusion has a significant effect in many important processes in nature and technology. Thermohaline convection in oceans, which is a salinity gradient caused by a temperature difference is due to this Soret effect [1]. In the analysis of the distribution of components in oil reservoirs, thermal diffusion together with isothermal and pressure diffusion are important [2]. Other applications include isotope separation in liquid and gaseous mixtures [3] and characterization and separation of polymers [4].

The temperature and concentration gradients in a binary system are coupled as a consequence of the Soret effect. This coupling results in a density gradient. Component separation describes the influence of thermal diffusion on the convection in a binary mixture. Precise measurements of the diffusion and Soret coefficients require repression of macroscopic flow in the mixtures. However, achieving this condition is quite difficult in terrestrial conditions because of thermal and solutal convection and double-diffusive instability. Space flight conditions also produce vibrations (g-jitters) caused by aerodynamic forces, non-ideal free falling and trembling of the space vehicle, onboard machinery, and the crew's activity, which can lead to significant macroscopic flows. This background g-jitter may alter the benefits of the microgravity environment. Thus a study of the effects of controlled vibrations on the measurements of the diffusion and Soret coefficients in liquid systems could be beneficial. This makes it important to perform theoretical and numerical studies of the influence of vibration on temperature and concentration fields in diffusive mixtures.

Although g-jitter seems to have a major impact on diffusion-related experiments in Space, very few experimental studies have addressed this topic. In these experiments, the temperature was monitored at several fixed points. The results did not provide clear evidence of time-averaged flows nor of the related heat transfer caused by periodic high-frequency g-jitter.

It is known that the effects of vibration on a fluid system with a density gradient resulting from a non-uniform temperature field are relative flows inside the fluid which are called thermovibrational convection. These relative motions result from the different inertia of the cold and hot parts of the fluid, which have different densities. Vibrations can also strengthen gravitational convection, which depends on the mutual orientation of the vibration axis and the thermal or the compositional gradient. A pure thermovibrational convective mechanism can be observed in weightlessness only as a result of eliminating static gravity that can provide an additional driving force for convective motion. The flow fields can be decomposed into the 'quick' part, which oscillates with the frequency of vibration, and the 'slow' time-averaged part (mean flow). The latter represents a non-linear response of the fluid to a periodic excitation. Vibrational convection provides a mechanism of heat and mass transfer as a result of mean flows. In weightlessness, this can be an additional method of transporting heat and matter [5].

The experiment IVIDIL (Influence of Vibration on Diffusion of Liquids) [44,46] examined the influence of vibration stimuli on diffusive phenomena. It was performed on a binary mixture of water and isopropanol at mass fractions of 0.9-0.1 and 0.5-0.5. Particularly for this mixture, the sign of the Soret coefficient depends on the concentration. As shown in Figure 1.1, it is positive  $(S_T > 0)$  if the water content is less than 75%, otherwise it is negative  $(S_T < 0)$ . Therefore, a wide variety of phenomena can be studied using the same mixture. In binary mixtures, the positive Soret effect is when the lighter component is driven towards the higher temperature region and the negative Soret effect when the situation is the opposite.



Figure 1.1: Soret coefficient in different compositions of water and isopropanol mixtures.

## **1.1** Thesis arrangement and objectives

The effect of vibration (g-jitter) and Soret effect was studied in a fluid mixture by performing a direct numerical analysis of the transient process using the finite volume method. A finite twodimensional model of a binary mixture of water and isopropanol (C3H7OH) is considered and subjected to a temperature gradient normal to the vibrational instability (g-jitter). The objective was to determine the dependence of the physical properties of the mixtures on time and concentration. Also, calculating thermal convection assuming the vibration has a sinusoidal translational form. In addition, the role of onboard g-jitters was discussed in detail by analyzing the experimental results and by comparing with the numerical calculations. The results helped to identify the limit level of vibrations where g-jitter does not play a role. This was satisfied by observing the diffusive process under different imposed and controlled vibrations. Moreover, vibration-induced convection and, particularly, heat and mass transfer under different levels of vibration investigated.

The theoretical concepts, studied papers, and relevant literature are discussed in Chapter two. The mathematical formulation of the problem in dimensional form, geometrical and numerical models and related boundary conditions are introduced in Chapter three. In Chapter four, two benchmark studies are compared and discussed in detail. Thermal diffusion without considering the Soret effect for two mixtures under the influence of the same vibrations in a zero-gravity environment is studied and benchmarked against the work of other scientific groups. The velocity line plots and instantaneous flow patterns are also compared. The second benchmark is similar to the first but under the influence of different levels of vibration. The mean flow patterns along with the related Nusselt numbers near the walls are compared with those of other groups. In Chapter five, the effects of variable parameters on the process under the influence of vibrational instability are studied in detail. The PC-SAFT equation of state and Boussinesq approximation are applied and evaluated. In Chapter six, various numerical results are compared with the results obtained from the experimental analysis. The final conclusion is made based on both experimental results and various numerical analyses. The general conclusions, contributions and possible future works are described in Chapter seven, and related suggestions and recommendations are discussed.

# **Chapter 2**

# **Theory and Literature Review**

## 2.1 Basic concepts in diffusion

## 2.1.1 Diffusion

Diffusion is the mechanism by which components of a mixture are transported in a mixture by means of random molecular (Brownian) motion. It can also be described as the process by which molecules spread from areas of high concentration to areas of low concentration. For example, diffusing molecules will move randomly between areas of high and low concentration, but because there are more molecules in the high concentration region, more molecules will leave the high concentration region than the low concentration one. Therefore, there will be a net movement of molecules from high to low concentration. Initially, a concentration gradient leaves a smooth decrease in concentration from high to low which will form between the two regions. As time progresses, the gradient will grow increasingly shallow until the concentrations are equalized.

Diffusion is a spontaneous process. It is simply the statistical outcome of random motion. Diffusion increases entropy and decreases Gibbs free energy, and therefore is thermodynamically favourable. Diffusion operates within the boundaries of the Second Law of Thermodynamics.

The diffusion equation provides a mathematical description of diffusion. This equation is derived from Fick's law, which states that the net movement of diffusing substance per cross-sectional area (the flux) is proportional to the concentration gradient and is therefore towards the area of lower concentration. Thus, if the concentration is uniform there will be no net motion. The constant of proportionality is the diffusion coefficient, which depends on the diffusing species and the material through which diffusion occurs. An analogous statement of Fick's law, for heat instead of concentration, is Fourier's law.

The mechanism of diffusion is "Brownian motion" whereby a molecule makes a random walk about a central location; since by kinetic theory the mean velocity of a particle is zero if it is not subject to any external forces. Due to collisions with neighbouring molecules, the motion of the particle is characterized by a mean free path which tends to confine the particle. But since there is no potential field acting to restore a particle to its original position, it is still free to move about the vessel or liquid in which it is located.

### 2.1.2 Fick's laws of diffusion

Fick's first law describes diffusion and can be used to solve for the diffusion coefficient ( $D_M$ ). It was derived by Adolf Fick in 1855. It relates the diffusive flux to the concentration field, by postulating that the flux goes from regions of high concentration to regions of low concentration, with a magnitude that is proportional to the concentration gradient. In one dimension, it can be written as:

$$J = -D_M \times \partial \rho / \partial x \tag{2.1}$$

where

- *J* is the diffusion flux in dimensions of [kg m<sup>-2</sup>s<sup>-1</sup>]; it measures the amount of substance that will flow through a small area during a small time interval.
- $D_M$  is the diffusion coefficient in dimensions of  $[m^2 s^{-1}]$
- $\rho$  is the concentration in dimensions of [kg m<sup>-3</sup>]
- *x* is the position [m]

 $D_M$  is proportional to the squared velocity of the diffusing particles, which depends on the temperature, viscosity of the fluid and the size of the particles.

Fick's second law predicts how diffusion causes the concentration field to change with time:

$$\partial \rho / \partial x = D_M \partial^2 \rho / \partial x^2 \tag{2.2}$$

#### **2.1.3 Diffusion equation**

The diffusion equation can be derived in a straightforward way from the continuity equation, which states that a change in density in any part of the system is due to inflow and outflow of

material into and out of that part of the system. Effectively, no material is created or destroyed:

$$\frac{\partial \rho}{\partial t} + \nabla . \vec{j} = 0 \tag{2.3}$$

where  $\vec{j}$  is the flux of the diffusing material. The diffusion equation can be obtained easily from this when combined with the phenomenological Fick's first law, which assumes that the flux of the diffusing material in any part of the system is proportional to the local density gradient:

$$\vec{j} = -D_M(\rho)\nabla\rho(\vec{r},t)$$
(2.4)

The diffusion equation is a partial differential equation which describes density fluctuations in a material undergoing diffusion. It is also used to describe processes exhibiting diffusive-like behaviour. The equation is usually written as:

$$\partial \rho / \partial t = \nabla \left( D_M(\rho, \vec{r}) \cdot \nabla \rho(\vec{r}, t) \right)$$
(2.5)

where  $\rho(\vec{r}, t)$  is the density of the diffusing material at location  $\vec{r}$  and time t and  $D_M(\rho, \vec{r})$  is the collective diffusion coefficient for density  $\rho$  at location  $\vec{r}$ . If the diffusion coefficient depends on the density then the equation is nonlinear, otherwise it is linear. If  $D_M$  is constant, then the equation reduces to the following linear equation:

$$\partial \rho / \partial t = D_M \nabla^2 \rho(\vec{r}, t) \tag{2.6}$$

#### 2.1.4 Molecular diffusion

Consider a water tank with a portion containing blue water and the other portion yellow water, separated by an impermeable membrane. If the membrane is suddenly removed, formation of a greenish mixing zone can be observed at the interface of the two portions. As time progresses, the width of the mixed zone grows. The rate of growth generally depends on the temperature of the fluid and its molecular weight. The growth of the zone implies an increase in mixing. The

cause of the mixing is attributed to the kinetic activity of molecules, which provides the "random" motion of the molecules leading to mixing. This mixing process is generally called "molecular diffusion". We can also call it "random motion" to reflect the fact that we cannot describe the motion of each individual molecule in a simple mathematical terms, and the fact that we are generally not interested in the behaviour of each individual molecule (i.e., microscopic behaviour) [6].

Diffusion or smearing is a macro scale phenomenon. It is a result of the observation of fluid behaviour averaged over a volume. In fact, at the microscopic scale, each blue molecule is still a blue molecule and each yellow molecule is still yellow. The change of colour simply is a reflection of the presence of different numbers of blue and yellow molecules in a given volume greater than the molecule itself.

From the water tank experiment, it can be observed that the blue fluid move into the yellow fluid where the blue is absent or yellow is dominant. Thus, it is logical to conclude that the diffusion takes place from a high concentration region to a low concentration region.

#### **2.1.5 Thermal diffusion**

Thermal diffusion is the movement of fluids, either gaseous or liquid, influenced by the temperature of the fluid. A temperature gradient applied to a liquid mixture not only causes a heat flux but also gives rise to a diffusion current of the constituent components. The resulting separation of the components causes a concentration gradient parallel or anti-parallel to the temperature gradient, as shown in Figure 2.1. This cross-effect between temperature and concentration is known as thermal diffusion or Ludwig-Soret effect. Since its discovery by Ludwig (1856) and the first systematic investigations in liquid mixtures by Soret (1879), the effect has been subject to experimental and theoretical studies. Furthermore, the effect is relevant in technical applications such as polymer characterization, the analysis of petroleum reservoirs and for accumulation of uranium.



**Figure 2.1:** Illustration of the Soret effect. If a temperature gradient is applied to a mixture, this results in a concentration gradient.

# 2.2 Approximation and Equation of State

## 2.2.1 Boussinesq approximation

Generally, in fluid dynamics, the Boussinesq approximation (named for Joseph Valentin Boussinesq) is used in the field of buoyancy-driven flow (also known as natural convection). Although these equations are named after Boussinesq, they seem to have been first used by Oberbeck. It states that density differences are sufficiently small to be neglected, except where they appear in terms multiplied by "g", the acceleration due to gravity. The essence of the Boussinesq approximation is that the difference in inertia is negligible but gravity is sufficiently strong to make the specific weight appreciably different between the two fluids.

The application of this knowledge to technological or environmental problems can be meaningful only when the conditions under which the Boussinesq equation is valid are explicitly known. This approximation is commonly understood to consist of the following:

- 1. Density is assumed constant except when it directly causes buoyant forces;
- 2. All other fluid properties are assumed constant;
- 3. Viscous dissipation is assumed negligible.

The first point means that the continuity equation has its incompressible form and that density is considered variable only in the gravitational term of the momentum equation. The other points simplify the equations so that attention is focused on the effects of buoyancy.

The Boussinesq approximation is inaccurate when the non-dimensionalised density difference is of order unity. On the other hand, the density difference between liquids  $\Delta \rho$  is assumed to be small, i.e.  $\Delta \rho / \rho_0 \ll 1$ .

In general, the Boussinesq approximation is for problems in which variations in temperature as well as variations in density are small. In these cases, the variations in volume expansion due to temperature gradients will also be small. The Boussinesq approximation is often used to simplify the equations governing fluid motion in order to facilitate both theoretical analysis and numerical computation. Therefore, it can simplify problems and save computational time. The basis of this approximation is that there are flows in which the temperature varies little, and therefore the density varies little, yet in which the buoyancy drives the motion. Thus the variation in density is neglected everywhere except in the buoyancy term.

#### 2.2.2 PC-SAFT Equation of State

The Statistical Associating Fluid Theory (SAFT) approach has been extensively applied to phase equilibrium since 1990. The perturbed-chain statistical associating fluid theory (PC-SAFT) equation of state, developed by Sadowski et al [7, 8], is an assay to model asymmetric and highly non-ideal systems and the results so far are rather favourable. It has been shown to give an improvement performance over the original SAFT equation of state.

In thermal diffusion models, equilibrium thermodynamic quantities are widely used. Therefore, it is a prerequisite to know reliable thermodynamic properties for the theoretical research on thermal diffusion. Consequently, for a certain mixture, the accuracy of the model relies not only on the particular model itself but also on the thermodynamic properties of the mixture. The thermodynamic properties cannot be obtained from the literature. As well, some are difficult to measure accurately. One reasonable approach to obtain the required thermodynamic properties is to use an equation of state. Here, a state-of-the-art equation of state PC-SAFT is used.

Thermodynamic properties such as the chemical potential, the residual partial molar internal energy, the partial molar enthalpy and partial molar energy as well as mixture density can be derived from the PC-SAFT equation of state.

The essence of PC-SAFT is to sum various contributions from the hard chain, dispersion, and association components of the residual Helmholtz energy:

$$\tilde{a}^{res} = \frac{A^{res}}{Nk_BT} = \tilde{a}^{hc} + \tilde{a}^{disp} + \tilde{a}^{assoc}$$
(2.7)

where  $A^{res}$  is the residual Helmholtz free energy, N is the total number of molecules,  $\tilde{a}^{hc}$  is the part of the Helmholtz energy from the hard chain contribution,  $\tilde{a}^{disp}$  is the part from the dispersive contribution, and  $\tilde{a}^{assoc}$  is the part from the association contribution. The hard chain contribution  $\tilde{a}^{hc}$  represents an expression for the reduced residual Helmholtz energy for homonuclear hard sphere chains. It consists of two parts: the hard sphere term and the chain term. The dispersive contribution  $\tilde{a}^{disp}$  is calculated from the second-order perturbation. After defining the reference chain fluid in the hard chain contribution, the dispersion contribution to the Helmholtz free energy is given as a summation of the first and second-order contributions. The extensive details of PC-SAFT can be found in References [7, 8].

### 2.3 Literature review

#### **2.3.1** Theoretical approach of thermal diffusion in different mixtures

Stenby et al. [9] test some thermodynamic models for the thermal diffusion factors of binary mixtures models in combination with different equations of state. The calculated values of thermal diffusion factors were compared with a few sets of experimental data for hydrocarbon mixtures. For calculation of the partial molar properties, they applied different thermodynamic models, such as the Soave–Redlich–Kwong and the Peng–Robinson equations of state. Their work was largely based on the work of Denbigh, who described in detail the thermodynamic approach to thermal diffusion based on the concept of the thermal heat of transport. They evaluated and compared some of the proposed models with experimental data.

Shukla and Firoozabadi [10] studied a new model for the prediction of thermal diffusion coefficients in binary mixtures of reservoir fluids using the thermodynamics of irreversible processes. They used equilibrium properties of mixtures and the energy of viscous flow. Equilibrium properties were obtained from the volume-translated Peng-Robinson equation of state, and the energy of viscous flow was estimated from viscosity. Their model can be used to predict thermal diffusion coefficients of several mixtures consisting of non-hydrocarbon and hydrocarbon fluids. Comparisons of theoretical results with experimental data shows a good performance of the model except in the near-critical region where all existing models are deficient. They also modified some of the earlier models, such as the Kempers, Haase, and Rutherford models, which are based on phenomenological and kinetic approaches. Additionally, they tested theoretical results against experimental data for the numerous hydrocarbon and non-hydrocarbon mixtures.

Later, Firoozabadi et al. [11] derived a theoretical model for thermal diffusion coefficients in ideal and non-ideal multi-component mixtures. Their model is based on the thermodynamics of irreversible processes and the molecular kinetic approach incorporating explicit effects of non-equilibrium properties, such as the net heat of transport and molecular diffusion coefficients, and of equilibrium properties of the mixture, which are determined by the Peng-Robinson equation of state. Their model successfully describes thermal diffusion factors of binary mixtures for which experimental data are available, even those in extreme non-ideal conditions and those close to the critical point. Their work demonstrated for the first time that there is no need to adopt a sign convention for thermal diffusion coefficients in binary and higher mixtures. The thermodynamic stability analysis showed that when the thermal diffusion coefficient is positive, the component should go to the cold region in a binary mixture. Their model shows the explicit dependency of the thermal diffusion factors in non-ideal mixtures of more than two components on molecular diffusion coefficients. The single validation of the model for a ternary mixture shows success.

Ghorayeb and Firoozabadi [12] derived a formalism based on powerful concepts of the thermodynamics of irreversible processes for calculating multi-component diffusion flux both at and away from the critical point. The derivations are based on the entropy balance expression combined with the phenomenological equations and Onsager reciprocal relations. The formalism

results in a clear expression of the thermal contribution in the diffusion flux for non-ideal multicomponent mixtures. The diffusion flux analysis at the critical point showed that, unlike isothermal and isobaric conditions where molecular diffusion flux is zero, molecular diffusion flux is finite and nonzero at the critical point. The thermal contribution in the diffusion flux is also finite at the critical point in multi-component mixtures. At non-isothermal conditions, as well as non-isothermal and non-isobaric conditions, the composition gradient reaches infinity at the critical point; therefore, the mole fraction plot vs. spatial coordinates has an inflection point. This formalism has allowed evaluation of thermal diffusion factors for ternary and higher mixtures that have not been attempted in the past.

Firoozabadi et al. [13] worked on the analysis of species separation in a thermo-gravitational column filled with porous media that so far has been based on strong dependency of thermal and molecular diffusion to dispersion. They suggest an alternative and show that the dispersion effect is negligible for the conditions in a packed thermo-gravitational column and that compositional dependency of the thermal diffusion should be accounted for. Their work shows that the thermal diffusion coefficient is a function of composition. Their results reveal that the dispersion effect may not be significant mainly due to low velocities. In their work, a new semi-implicit code is developed to perform computations efficiently. Their model shows good agreement with experimental data for both the binary mixture and the ternary mixture. This could be due to the proper prediction of thermal and molecular diffusion coefficients or the proper prediction of composition coefficients.

Haugen and Firoozabadi [14] developed the theory for using the deflection of laser beams of various wavelengths to determine molecular and thermal diffusion coefficients in multicomponent mixtures. They determine thermal and molecular diffusion coefficients of multicomponent mixtures from transient measurements of the deflections of laser beams of different wavelengths passing through a thermal diffusion cell. In the past, simultaneous determinations of molecular and thermal diffusion coefficients have only been achieved for binary mixtures. Their procedure is faster and more accurate than the current techniques to determine molecular diffusion coefficients. They show in an example that the molecular and thermal diffusion coefficients can be determined reliably in a ternary mixture. Their derivations are based on the assumption that thermal diffusivity is much greater than molecular diffusivity. This is generally true for liquids but not for gases. The practical limitations of their method depend on the noise in the measurements and the sampling frequency.

Dios and Firoozabadi [15] studied new molecular and thermal diffusion coefficients of binary mixtures of normal decane - normal alkanes and methylnaphthalene - normal alkanes, measured at atmospheric pressure and T=25°C. They measured thermal diffusion coefficients in a thermogravitational column. Also, molecular diffusion coefficients were measured using an open-ended capillary tube technique. Their results show a significant effect of molecular shape and size on thermal and molecular diffusion coefficients. Molecular diffusion coefficients show a monotonic behaviour in both aromatic - normal alkane and normal decane - normal alkane mixtures. Thermal diffusion coefficients reveal a non-monotonic trend with molecular size in the normal decane - normal alkane mixtures. Their report is the first discussing non-monotonic behaviour in the literature. They provided an accurate self-molecular diffusion coefficient for n-C<sub>10</sub> from binary data. They provide a systematic set of new measurements for two drastically different sets of binary hydrocarbon mixtures.

Kempers [16] worked on the calculation of the magnitude of the cross-effect, known as the Hertz effect, which is possible if all coefficients of the diffusion tensor of the mixture are known. For dilute gases, kinetic gas theory enables the calculation of the diffusion tensor, but calculation methods for dense gases, liquids, and solids require the input of measured binary or tracer diffusion coefficients. In this paper, for the first time, a calculation method for the magnitude of the Hertz effect without using input of any measured diffusion coefficient is presented. Input is required from the equation-of-state of the mixture and from kinetic gas theory. The theory is based on the principle of a maximum number of possible microstates in the stationary, non-equilibrium state. The theory has been compared with all of the measured diffusion data of mixtures. There is fair to good agreement between measurement and theory, but the Hertz effect in these mixtures is small.

Gross and Sadowski [7] modified the SAFT equation of state by applying the perturbation theory of Barker and Henderson to a hard-chain reference fluid. With conventional one-fluid mixing

rules, the equation of state is applicable to mixtures of small spherical molecules such as gases, non-spherical solvents, and chainlike polymers. The three pure-component parameters required for non-associating molecules were identified for 78 substances by correlating vapour pressures and liquid volumes. The equation of state gives good fits to these properties and agrees well with caloric properties. They compare it with the SAFT version of Huang and Radosz and reveal a clear improvement of the proposed model. Also they compare it with the Peng-Robinson model, confirming good performance of the suggested equation of state.

Gross and Sadowski [8] studied the application of the PC-SAFT equation of state to associating systems. Modeling the phase equilibrium and thermodynamic properties of systems in which molecules exhibit associating interactions remains a challenging problem in the chemical industry. The large number of recent studies on equations of state applied to associating systems is an indication of this circumstance. The PC-SAFT equation of state is applied to pure associating components as well as to vapour-liquid and liquid-liquid equilibrium of binary mixtures of associating substances. For these substances, the PC-SAFT equation of state requires five pure component parameters, two of which characterize the association. The pure-component parameters were identified for 18 associating substances by correlating vapour pressure and liquid density data. A comparison to an earlier version of SAFT confirms the good results for pure substances. Both vapour-liquid and liquid-liquid equilibrium were calculated for mixtures of this type using one constant  $k_{ij}$  parameter that corrects the dispersive interactions. Liquid-liquid and vapour-liquid equilibrium were also calculated for mixtures with two associating substances. Simple combination rules were adopted for the cross-associating interactions in this study; no additional binary interaction parameter was thereby introduced. A constant  $k_{ii}$  parameter correcting the contribution due to dispersion was used in the correlation of vapour-liquid and liquid-liquid equilibrium.

Jamet et al. [17] studied two methods for measuring the Soret coefficient of water – alcohol mixtures (10% mass fraction of alcohol: ethanol or propanol). Their methods, performed with porous packing, are particularly adapted for apparent negative Soret coefficient determination. Their first method uses a flat-packed Soret cell operated under a downwards vertical thermal gradient, and the second method uses a packed cylindrical thermal diffusion column and thermo-

gravitation. They used experimental and modeling results to estimate the Soret coefficient, from results for the two different packed cells. Finally, they compared the results with numerical results from the METSOR code.

Strathmann et al [18] studied Soret coefficients of the ternary system of poly ethylene oxide in mixed water - ethanol solvents over a wide solvent composition range by means of thermal diffusion forced Rayleigh scattering. The Soret coefficient  $S_T$  of the polymer was found to change sign as the water content of the solvent increases with the sign change taking place at a water mass fraction of 0.83 at a temperature of 22 °C. For high water concentrations, the value of  $S_T$  of poly ethylene oxide is positive, i.e., the polymer migrates to the cooler regions of the fluid, as is typical for polymers in good solvents. For low water content, on the other hand, the Soret coefficient of the polymer is negative, i.e., the polymer migrates to the warmer regions of the fluid. Measurements for two different polymer concentrations showed a larger magnitude of the Soret coefficient for a smaller polymer concentration. The temperature dependence of the Soret coefficient was investigated for water-rich polymer solutions and revealed a sign change from negative to positive as the temperature is increased. They also performed thermal diffusion experiments on the binary mixture water - ethanol. For the binary mixtures, the Soret coefficient of water was observed to change sign at a water mass fraction of 0.71. This is in agreement with experimental results from the literature. Their results show that specific interactions hydrogen bonds between solvent molecules and between polymer and solvent molecules play an important role in thermal diffusion.

Firoozabadi et al. [19] studied thermal diffusion coefficients in three ternary mixtures that were measured in a thermo-gravitational column. One of the mixtures consisted of one normal alkane and two aromatics dodecane – isobutyl benzene – tetra hydro naphthalene, and the other two consisted of two normal alkanes and one aromatic octane – decane - 1-methylnaphthalene. The results for ternary mixtures of octane – decane - 1-methylnaphthalene show a sign change of the thermal diffusion coefficient as the decane composition changes, despite the fact that the two normal alkanes are similar. In addition to thermal diffusion coefficients, molecular diffusion coefficients were also measured for three binary mixtures and one of the ternary mixtures. They used the open-end capillary-tube method in the measurement of molecular diffusion coefficients.
The molecular and thermal diffusion coefficients allow the estimation of thermal diffusion factors in binary and ternary mixtures. However, in the ternary mixtures one also has to calculate phenomenological coefficients from the molecular diffusion coefficients. They also did a comparison of the binary and ternary thermal diffusion factors for the mixtures, which revealed a remarkable difference in the thermal diffusion behaviour in the binary and ternary mixtures. Their results showed that the binary and ternary thermal diffusion factors for the same species are very different. The comparison of the measured thermal diffusion coefficients of the binary mixtures *n*-C<sub>8</sub>-methyl naphthalene and *n*-C<sub>10</sub>-methyl naphthalene and ternary mixture. In the ternary mixture, the thermal diffusion coefficient of *n*-C<sub>10</sub> changes sign as the concentration of the mixture changes.

Sengers et al. [20] studied how a temperature gradient in a liquid mixture causes a concentration gradient through the Soret effect. They developed an instrument to measure the Soret effect by observing the bending of a laser beam propagating horizontally through the liquid mixture subjected to a temperature gradient in the vertical direction. Their design of the liquid cell, with a long path length and controlled temperature uniformity, enables them to measure Soret coefficients with an accuracy of 1-3 %, higher than that obtained by previous investigators. In addition, by measuring the dynamic response of the beam deflection after imposition of the temperature gradient, they also determine the mutual diffusion coefficient. They have applied the technique to mixtures of toluene and n-hexane over the temperature range 5-45 °C and to mixtures of ethanol and water at 25 °C. They have verified that the measured value of the Soret coefficient is independent of the magnitude of the temperature gradient imposed up to 14 K/cm. The Soret coefficients obtained for mixtures of toluene and *n*-hexane differ from the values obtained by previous investigators with a thermo-gravitational column method, but they are in good agreement with the results obtained by Kohler and Muller with a forced Rayleigh scattering method. For mixtures of ethanol and water, their Soret coefficients agree with the results obtained earlier by Kolodner et al., who also used an optical beam-bending technique.

Pan et al. [21] studied the theoretical approach to evaluate thermal diffusion in aqueous solutions. While the thermal diffusion in gases has been widely investigated and can be well

explained by kinetic theory, the thermal diffusion in liquids has also raised increasing scientific and engineering interest in recent years. Flow due to thermal diffusion may change direction in fluid mixtures with the variation of composition and temperature. This event is of great interest in petroleum research. Using a modified theoretical approach, they evaluate the thermal diffusion factor in alcohol water mixtures, including methanol, ethanol, and isopropanol aqueous mixtures. By combining this approach with a PS-SAFT equation of state and using two adjustable parameters calculated from experimental data, they obtained good agreement for the prediction of thermal diffusion in alcohol water mixtures when compared with the available experimental data. The essence of their approach is to express the thermal diffusion factor in terms of thermodynamic properties, which can be obtained by using PC-SAFT. The predicted thermal diffusion factors agreed well with the available experimental data. The results also suggest the significance of thermal diffusion in infinite dilutions in thermal diffusion research.

Pan et al. [22] studied thermal diffusion in water – methanol, water – ethanol, and water - isopropanol mixtures using the PC-SAFT equation of state in a theoretical prediction of thermal diffusion. They also optimized the parameters of the PC-SAFT for these mixtures. They used several thermal diffusion models available in the literature with the thermodynamic properties from the PC-SAFT EOS. The Firoozabadi model combined with the PC-SAFT EOS has shown an effective capability for predicting mixtures with a low to moderate water concentration. They show that PC-SAFT equation of state is capable of predicting thermodynamic properties for water–alcohol systems like water – methanol, water – ethanol, and water – isopropanol.

#### **2.3.2** Vibration effects on diffusion in a reduced gravity environment

Lyubimova et al. [23] numerically investigated the effect of static and vibrational acceleration on the measurement of diffusion and Soret coefficients in binary mixtures under low gravity conditions. High-frequency vibration allows the decomposition of variables into slowly varying (time-averaged) and quickly oscillating (pulsational) components. The effect of vibrations applied in two directions, perpendicular and parallel to the imposed temperature gradient was investigated by numerical simulation. The study was performed for both zero gravity and low gravity conditions. In the latter case, a residual flow would exist which can be damped by applying an appropriate vibration intensity in the appropriate direction. Melnikov et al. [24] studied the impact of two types of vibrations (sine and cosine) at the onset of thermovibrational convective flow. The development of thermovibrational convection in a cubic cell filled with isopropanol was investigated. It was shown that the initial vibrational phase plays a significant role in the transient behaviour of thermovibrational convective flow. It was found that under the cosine vibrations, the flow reaches the steady state two times slower than under the sine vibrations, and the Nusselt number is almost ten times smaller. The flow and temperature fields were also investigated and compared in the two cases. It was shown that starting with sine accelerations causes significantly larger initial transport than if the accelerations start as a cosine function.

Shevtsova et al. [25] studied the thermoconvective flows in an experimental test of convection caused by translational vibration of a non-uniformly heated fluid in low gravity. The temperature fields were observed from the examined cubic cell and studied in a wide range of frequencies and amplitudes. It was confirmed that vibrational convection intensifies the heat transfer in the system. This transport was found to be significantly weaker in the absence of vibration and negligibly small in normal gravity for the studied levels of vibration. In addition, by utilizing particle tracing in the experiment, the previous mean flow structures from the numerical studies were verified.

Shevtsova et al. [26] analyzed in detail the residual accelerations of the Foton-12 mission. The collected data evaluated from different accelerometers showed that the residual gravity with small amplitude and low frequency vibration exists due to atmospheric drag, the stabilizing rotation, and the complex motion of the platform along the orbital trajectory.

Yan et al. [27] performed a numerical study on the effect of low frequency g-jitter and static gravity on thermal diffusion. It was shown that the overall effect of vibrations on diffusion can be related to a nonlinear interaction between the effects caused by each individual g-jitter component. In the presence of large static gravity, the Soret separation is overshadowed by the convection in the mixture. In other words, the accuracy of the diffusion measurement will be affected significantly. The importance of periodic vibration on diffusion depends on the frequency. Low frequency vibration has more effect on diffusion measurements compared to

high frequency vibration. It was also found that when both static residual gravity and the oscillatory g-jitter component in the direction perpendicular to the temperature gradient exist, the flow field is intensified

Yan et al. [28] studied the thermal diffusion process under different microgravity environments using measured g-jitter data from onboard the International Space Station (ISS) and FOTON-12. It was found that the diffusion process is only mildly affected by the g-jitters in both platforms. It was concluded that a proper choice of experimental location is of great importance in minimizing the effect of g-jitters. It was also shown that the undesirable effect of large oscillatory g-jitters similar to the quasi steady residual-g should be controlled. However, it is important to indicate that the data (g-jitters) collected from the International Space Station (ISS) were obtained when the ISS was not in full assembly.

Yan et al. [29] studied the effect of g-jitter on thermal diffusion on-board the International Space Station. It was found that the ISS microgravity environment can disturb the diffusion process from the ideal condition in some circumstances depending on the magnitude of the g-jitter. It was also shown that frequency and alignment of the experimental cell are both equally important.

Melnikov et al. [30] performed numerical and experimental investigation in preparation of a parabolic flight experiment. The work describes the preparation of experiments on thermovibrational convection in microgravity during parabolic flights. It describes the experimental setup for observing thermovibrational flows. It consists of two principle parts: a transparent cubic cell filled with liquid and subjected to harmonic vibrations with a help of linear motor, and an optical interferometer coupled with a digital recorder and processer for observing velocity and temperature fields in the cell. The working liquid and control parameters of the experiment were investigated in a 3-D numerical simulation of thermovibrational convection in a cubic cavity for real parabolic flight conditions. The study was aimed at estimating the values of physical quantities that manifest in the presence of thermovibrational flows and can be experimentally measured during short microgravity time (20 s). They have formulated the requirements, which should be satisfactory for successful observation of thermovibrational convectional convection in the microgravity of a parabolic flight. Based on these requirements, a strategy for choosing the working liquid and control parameters of the experiment was proposed. It was

found that among several liquids considered, isopropanol is the best choice due to its large thermal expansion and small viscous time, which is important due to the short duration of microgravity (20 s). Their numerical modelling of the experiment shown that the proper choice of parameters allows obtaining thermovibrational flows which are strong enough to be experimentally observed and recorded by the available equipment.

Kerr and Zebib [31] examined the double-diffusive convection in a liquid layer under highfrequency vibrations. They looked at the higher order asymptotic wave numbers and determined how vibration affects the stability boundary in the region of maximum instability. He found that the stability boundaries for the non-vibrating case and the predictions of the small asymptotes would all be essentially independent of the mass/thermal diffusivity ratio and Prandtl number. This indicates the central role of mass diffusion in the instabilities that were observed. Razi et al. [32] considered the influence of vibrations on the stability threshold of two-dimensional Soretdriven convection in the high frequency and small amplitude cases. Their results showed that increasing the direction of vibration has a stabilizing effect on the quasi-equilibrium solution and decreases the oscillatory frequency.

The effect of average vibrational convection produced by high-frequency vibrations has been considered by Gershuni et al. [33, 34] in a binary mixture in the presence of Soret effect. The analysis shows interesting distinctive features. Two flow structures were observed: In the main regime, four-vortex behaviour, and in secondary regime one-vortex behaviour. The dependence of the dimensionless heat flux (Nusselt number) on the vibrational Rayleigh number or Gershuni number is presented for the two possible flow structures. It was also found that the structures are very sensitive to the variations of the Prandtl number.

Savino and Monti [35] studied the diffusion process in isothermal condition and a detailed analysis of vibrational impact on fluid physics experiments onboard microgravity platforms. It was shown that convective disturbances are related to both the amplitude and frequency of vibrations, as well as to the mutual orientation of the density gradient and the acceleration vector. They indicated that significant differences are found in the flow field arrangements and the time profiles of the thermofluid dynamic distortions in both of the two cases of natural convection: "residual-g" and vibrational convection "g-jitters".

Thermal diffusion in a binary mixture of methane and *n*-butane subject to g-jitters was investigated by Chacha et al. [36]. They found that the g-jitter produces mixing and changes the Soret effect in the cavity. The flow showed synchronous response to the oscillatory accelerations. Later, Chacha and Saghir [37] studied the effect of the time-dependent vertical gravity vector in a rectangular cell on the mass diffusion in a binary fluid mixture subject to a lateral temperature gradient. The effect of varying diffusion coefficients with the temperature and the fluid composition were analysed and compared to the average constant values. The numerical study shows the reduction of the compositional variation as a result of the increased convection produced by the g-jitter. They also found that the Soret coefficient oscillates with time at the same frequency as the original excitation. However, the backflow produced some disturbances which makes it non-sinusoidal in shape. Previous experiments onboard the MIR station using the Canadian MIM (Microgravity-vibration Isolation Mount) platform conducted by Smith et al. [38] showed that the diffusion coefficients in metals and alloys depend on the residual accelerations and the quality of microgravity.

Ryzhkov and Shevtsova [39] studied the thermal diffusion and convection in multi-component mixtures. They revised and extended the theoretical framework for describing the multi-component mixtures with the Soret effect. The separation ratio, a fundamental parameter characterizing the influence of thermal diffusion on convective phenomena, was generalized to the multi-component case. It was shown how to define this parameter for a particular component of the mixture. To characterize the multi-component system as a whole, the net separation ratio, which does not depend on the choice of solvent, is introduced. Based on these results, the dimensionless equations for convection in multicomponent mixture were derived. The proposed formulation was used in analyzing the steady state separation in a thermogravitational column. The distributions of velocity, temperature, and composition in a multi-component system were discussed in detail.

Shevtsova et al. [40] studied the double diffusive and vibrational convection with the Soret effect in a cubic rigid cell filled with 90% water and 10% isopropanol that was subjected to a temperature difference between opposite lateral walls. Numerical simulations were carried out for g-jitter induced flow. The direction of g-jitter was the same as the residual gravity vector, which was perpendicular to the applied temperature gradient. Various combinations of static and oscillatory components were used and vibrations with two different frequencies were examined: when the period of oscillations is smaller than any characteristic time i.e. viscous, thermal, and diffusion times, and when the period of oscillation is comparable with viscous time. Component separation due to the Soret effect under these driving actions was analyzed. The concept of timeaveraged models was used for the explanation of the high-frequency results. The interplay between the mean and fluctuating motions was discussed. Three research groups performed the benchmark of numerical solutions in three-dimensional Navier-Stokes, energy, and concentration equations using the true physical parameters for the space experiment. The objective was to carry out an accurate study of heat and mass transfer in a binary liquid mixture with the Soret effect in the presence of steady residual gravity and its oscillatory component, as well as to carry out a benchmark of numerical solutions. It was found that for the considered range of frequencies, the heat and mass transport increased with a decrease of frequency. Also, it was concluded that in the combined case of simultaneous static gravity and high-frequency g-jitter, the buoyancy determines the Soret separation. However, the nonlinear interaction of static and vibrational actions results in larger mean fields of the temperature and concentration than in the pure oscillatory case at the same frequency.

Mialdun and Shevtsova [41] developed an experimental technique based on optical digital interferometry for measuring thermo-diffusion (Soret) coefficients. The experiments were performed in a cubic cell containing an aqueous solution and were heated from above. This is the first report on the method that allows recording the temperature and concentration fields in the entire cell rather than just between two points. The time-dependent evolution of the concentration field allows the measurement of the molecular diffusion coefficient in addition to the Soret coefficients. The measurements of transport coefficients were performed in water/ethanol and water/isopropanol mixtures. All methods developed to measure thermal diffusion coefficients were affected by convection to one extent or another. Unlike previously

developed methods this method allows one to see and analyze the situation inside the cell. Their method has undergone extensive scientific validation against recently published data for water/ethanol mixtures. Also, their method was applied to the measurements of the Soret coefficients in water/isopropanol mixtures. Their experimental method was initially developed for future experiments in microgravity where buoyant convection is negligibly small. However, they showed that it can be used in laboratory conditions even when parts of the cell are affected by convection.

Mialdun et al. [42] studied the experimental evidence of convection caused by translational vibration of non-uniformly heated fluid in a low gravity environment. Previously, when the theory of vibrational convection in weightlessness was studied, direct experimental proof of the results had been missing. The authors performed a set of experiments that observe a temperature field in the front and side views of the cubic cell. They also employed particle tracing to study the flow. The evolution of the flow was studied systematically over a wide range of frequencies and amplitudes. The flow structures reported in their previous numerical studies were confirmed in these experiments. The transition from four-vortex flow to the pattern with three vortices is observed in the transient state.

Shevtsova et al. [43] studied the development of convection in a binary mixture with the Soret effect. The Space experiment IVIDIL studied the influence of vibration stimuli on the measurement of diffusion and thermal diffusion coefficients. A binary mixture of water and isopropanol was selected as the working liquid. The principle of the Space experiment and ground based research were discussed. Results of 3D numerical simulations of thermal vibrational convection in binary mixtures at the conditions of the parabolic flight were presented. Heat and mass transport arising in the system with an increase in vibration was discussed. It was found that periodic oscillations normal to the density gradient significantly increase the heat and mass transfer when the periodic oscillations are stronger than the static gravity. It was shown that non-linear interactions between the static and vibrational accelerations result in additional steady mean fields.

Shevtsova [44] highlighted the objectives of IVIDIL experiment as well as the experimental setup used. Numerical investigations prior to the experiment were briefly discussed. It was concluded that examining and studying the effect of controlled vibrations (i.e. with desired frequency and amplitude) can provide valuable quantitative information on how vibrations affect the measurements of different physical quantities in Space experiments. The objectives of the IVIDIL experiment were highlighted as: Understanding the kinetic mechanisms that drive diffusion in the presence of vibrations, analyzing the effect of g-jitters on molecular and thermal diffusion processes, identifying the threshold of vibrations below which g-jitters do not affect onboard experiments, interpreting previous Space experiments and new data obtained from the flight, investigating flow structures and heat transfer induced by thermal and solute vibrational convection, and providing a rationale in the definition of future experiments.

Mialdun and Shevtsova [45] discussed the digital interferometry tool used in studying the thermal diffusion effect. Interferometry is an optical technique for measurements of the refractivity of objects, from which related quantities such as temperature or concentration can be determined. It is considered the most precise method for measuring diffusion in transparent fluids. The authors discussed the main factors affecting the accuracy of thermal diffusion measurement by interferometry. For the measurements of thermal diffusion by means of optical diagnostics, they used a classical Soret cell with transparent lateral walls clamped between two thermo-stabilized blocks. Although this system is generally stable with a downward temperature gradient, some small lateral heat flux provokes residual convection which in turn can disturb the measurement. The ability of the method to locate and exclude from consideration the regions disturbed by convection was considered for cells of different geometries. They optimized the cell design with the help of digital interferometry to measure Soret and diffusion coefficients in different binary mixtures. The applicability, reliability and accuracy range of the method in measurement of the diffusion and thermal diffusion coefficients in single- or two-step experiments was presented in this paper.

Shevtsova et al. [46] discussed the IVIDIL experiment which was successfully completed on the ISS in January 2010. In this paper they focused on one of the objectives of the experiment: performing precise measurements of diffusion and thermal diffusion coefficients for binary

mixtures in the absence of gravity. They reported the first results of measurements of mass transport coefficients in a mixture with a negative Soret effect: 10% isopropanol (IPA) – 90% water. A general description of the ISS facility related to the experiment was briefly presented, as well as the description of the IVIDIL instrument and the successive steps of the experimental procedure. After comparing numerical and experimental results, as well as thoroughly analyzing the imperfections of the measurements, they concluded that the ISS results are fully viable. Guidelines were drawn for justification of the results in favour of the Space experiment.

Mialdun and Shevtsova [47] studied the development of the optical digital interferometry (ODI) technique for measuring diffusion and Soret coefficients. The feature of this method is that it traces the transient path of the system in the entire cross section of a cell. In this way, it is applicable not only for measurements of Soret coefficients, but also for studying diffusive transport and convection mechanisms. This method is not widely used so far and, in their view, not because of fundamental limitations but rather due to a lack of properly developed post-experimental procedures of raw data analysis. Thus, in this paper, their attention was focused on the successive analysis of different steps: the fringe analysis, the choice of reference images, the thermal design of the cell, and the multi-parameter fitting procedure. Using the ODI technique, they measured the diffusion and the Soret coefficients for the three benchmark systems composed of  $C_{12}H_{26}$ , IBB, and THN at a mean temperature of 25°C and 50 wt. % in each component. The results showed good overall agreement with the proposed benchmark references. In conclusion, their results support the general trend of other studies and prove that the suggested technique is advantageous in some applications.

Shevtsova et al. [48] discussed the experimental and theoretical study of vibration-induced thermal convection in low gravity. In this paper, an experimental and computational study of thermal vibrational convection in a reduced-gravity environment of a parabolic flight was performed. The transient evolution of the temperature field in a cubic cell subjected to translational vibration was investigated by optical digital interferometry. The mean flow structures previously reported in numerical studies were confirmed. The transition from four-vortex flow to a pattern with a large diagonal vortex and two small vortices was observed in the transient state. The experiments show a significant enhancement of heat transfer by vibrational

mean flows with increasing vibrational strength. They performed a three-dimensional direct numerical simulation with a real microgravity profile and two-dimensional numerical modelling based on an averaging approach. They presented comparisons with the experimental results. The influence of residual gravity on heat transfer was investigated numerically and correlated with the experimental data. It was found that gravity affects non-uniformly heated fluids and that this effect can be reproduced in weightlessness by applying vibrations to the system.

# **Chapter 3**

# Governing Equations, Geometrical Model, Boundary Conditions and Numerical Approaches

## **3.1 Governing equations**

A complete set of governing equations have been used to simulate thermo-solutal diffusion and convection. It consists of the mass conservation equation, momentum equation, energy equation and mass transfer equation. In addition, the PC-SAFT EOS [7, 8] and the mixing rule were used for calculating the density in the in-house code and FLUENT, respectively. The solved equations are as follows:

## 3.1.1 Mass conservation equation

The fluid comprises two components, isopropanol with a mass fraction of *C* and the carrier fluid, water, with a concentration being equal to  $C_0 = I - C$ . The following equation is used for mass conservation:

$$\frac{\partial\rho}{\partial t} + \frac{\partial\rho u}{\partial x} + \frac{\partial\rho v}{\partial y} = 0$$
(3.1)

where  $\rho$  is the density, *u* and *v* are the velocity component in the *x* and *y* directions, respectively, and *t* marks the time. The mass transfer equation for each component has the following form:

$$\rho\left(\frac{\partial c}{\partial t} + u\frac{\partial c}{\partial x} + v\frac{\partial c}{\partial y}\right) = -\vec{\nabla}.\vec{j}$$
(3.2)

where the total flux  $\vec{j}$  is calculated by the following equation which consists of the mass diffusive flux in the first term and the thermal diffusion flux in the second term.

$$\vec{j} = -\rho \left[ D_C \vec{\nabla} c + D_T \vec{\nabla} T \right]$$
(3.3)

The terms  $D_C$  and  $D_T$  are the mass diffusion coefficient and the thermal diffusion coefficient, respectively. They are assumed to either vary in terms of the local temperature and composition by using PC-SAFT EOS model in the in-house code or remain constant in the FLUENT.

#### 3.1.2 Momentum conservation equation

The Navier–Stokes equations are used in order to solve the velocity field. The equation in the x direction is written as:

$$\rho \left[ \frac{\partial u}{\partial t} + u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y} \right] = -\frac{\partial p}{\partial x} + \mu \left[ \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} \right] + \rho g_x$$
(3.4)

and in the y direction:

$$\rho \left[ \frac{\partial v}{\partial t} + u \frac{\partial v}{\partial x} + v \frac{\partial v}{\partial y} \right] = -\frac{\partial p}{\partial y} + \mu \left[ \frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial y^2} \right]$$
(3.5)

where

$$g_x = -g_{vib}\omega^2 Cos(\omega t)$$
 and  $\omega = 2\pi f$  (3.6)

The vibration is applied only in the x direction. The residual gravity is set equal to zero, while  $g_{vib}$  is the amplitude of the applied oscillating part (g-jitter) and f is the frequency of the oscillation ( $\omega$  is angular frequency). The pressure is marked by p and  $\mu$  is the dynamic viscosity of the mixture. The mixture dynamic viscosity  $\mu$  at each node is computed based on a simple mass fraction average of the pure species viscosities:

$$\mu = \sum_{i} C_{i} \mu_{i} \tag{3.7}$$

where  $C_i$  is the mass fraction of each component at each node and  $\mu_i$  is the pure component viscosity.

#### **3.1.3** Energy conservation equation

Assuming no internal heat source, the conservation of thermal energy is written as follows:

$$\rho c_p \left[ \frac{\partial T}{\partial t} + u \frac{\partial T}{\partial x} + v \frac{\partial T}{\partial y} \right] = k \left[ \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} \right]$$
(3.8)

where  $c_p$  is the specific heat, k is the thermal conductivity and T is the temperature of the fluid.

#### 3.1.4 Equation of state

It is necessary to define the relationship of the density in the fluid to the flow parameters such as temperature, pressure, and species concentration which is of the form  $f(\rho, p, T, c) = 0$  in order to make use of the preceding conservation equations. The state of the fluid (liquid state) is fixed by the initial composition, temperature, and pressure level. Small pressure variations in the cavity negligibly affect the density, and are therefore ignored. The main parameter governing the density variation in the process is the temperature variation; of a lesser but not negligible importance is the mixture compositional variation.

In the in-house code, the PC-SAFT equation of state [7, 8] is used to determine the density of the mixture as well as the fugacity which is needed to estimate the diffusion coefficients. The calculation of these properties were done at each node of the grid and re-calculated for each time step. In FLUENT, the density is calculated using the mixing rule when density is considered only as a function of concentration,

$$\rho = \frac{1}{\sum_{i} \frac{C_i}{\rho_i}} \tag{3.9}$$

where  $C_i$  is the mass fraction of the components at each node and  $\rho_i$  is the density of each component *i*. When the commercial code FLUENT is adopted, the density is also assumed to follow the Boussinesq approximation as shown in Equation 3.10.

$$\rho = \rho_0 [1 - \beta_T (T - T_0) + \beta_c (C - C_0)]$$
(3.10)

where  $\rho_0$  is evaluated using equation 3.9,  $\beta_c$  is the solutal expansion coefficient and  $\beta_T$  is the thermal expansion coefficient.

## **3.2** Geometrical model and boundary conditions

The physical model of the cavity under study is sketched in Figure 3.1.



Figure 3.1: Geometry and coordinate system. The gray vertical cross section is the plane where the analysis was conducted.

The system consists of a square cavity with a length of 0.01 m, containing the binary mixture of water and the isopropanol. The cavity is subject to the oscillatory component of the gravity called g-jitter which is modelled by a harmonic contribution to the body force. It is considered perpendicular to the thermal gradient and coincides with the *X* axis. The opposite walls are kept at constant temperatures  $T_{\text{hot}}$  and  $T_{\text{cold}}$  so the temperature difference is  $\Delta T = T_{\text{hot}} - T_{\text{cold}}$ . All other walls are assumed either thermally insulated or subject to a linear temperature profile, also rigid and impermeable. The applied boundary conditions are shown in Figure 3.1. The boundary conditions are thus applied as follows:

· Velocity boundary conditions

$$u = v = 0$$
 at  $x = 0$ ,  $x = L$ ,  $y = 0$  and  $y = L$  (3.11)

• Species boundary conditions

$$-D_C \frac{\partial c}{\partial x} + D_T \frac{\partial T}{\partial x} = 0 \quad at \quad x = 0 \quad and \quad x = L$$
(3.12)

$$-D_C \frac{\partial c}{\partial y} + D_T \frac{\partial T}{\partial y} = 0 \quad at \quad y = 0 \quad and \quad y = L$$
(3.13)

• Temperature boundary conditions

$$T_{hot} \quad at \quad y = L \qquad \& \qquad T_{cold} \quad at \quad y = 0 \tag{3.14}$$

## 3.3 Numerical approach and convergence criteria

In this analysis, continuity, mass, momentum and energy equations are discretized by the finite volume method and the SIMPLE algorithm [49] used for solving the continuity equation. In the case of the commercial software FLUENT, a second order Upwind discretization scheme was adopted for momentum and energy when PRESTO was used to describe the pressure. These selections were based on the recommendations in the FLUENT user's guide. PRESTO is recommended for the pressure-based solver and all types of meshes. Also, it is highly recommended for the high-Rayleigh-number natural convection problems. The SIMPLE method was used for pressure-velocity coupling and under-relaxation parameters for pressure, density, body forces, momentum and energy were set at 0.3, 1, 1, 0.7 and 1, respectively. These are the default values and in FLUENT, the default under-relaxation parameters for all variables are set to values that are near optimal for the largest possible number of cases. It was suggested to begin a calculation using the default under-relaxation factors. If the residuals continue to increase after the first 4 or 5 iterations, the under-relaxation factors should reduced. For most flows, the default under-relaxation factors do not usually require modification. If unstable or divergent behavior is

observed, however, we need to reduce the under relaxation factors. The convergence criteria were fixed at  $10^{-6}$  for continuity, velocities, and energy.

In addition, primitive variables (u, v, p, T) were employed. Afterwards, they are used to correct the velocity and pressure:

$$p = p^* + p'$$
 ,  $\vec{v} = \vec{v}^* + \vec{v}'$  (3.15)

In the above correlation equation,  $p^*$  is the initial estimate for pressure and is corrected by p'. Correspondingly,  $v^*$  is the related velocity obtained from the momentum equation. It is then corrected by v' achieved from following equations:

$$\vec{v}' = -K \cdot \nabla p'$$
,  $K = \begin{pmatrix} K_x & 0 & 0\\ 0 & K_y & 0\\ 0 & 0 & K_z \end{pmatrix}$  (3.16)

where K is a diagonal tensor resulting from the discretization scheme. The process continues until it meets the required convergence criteria. As a result of the strong coupling problem, the density and diffusion coefficients are calculated in terms of local temperature, pressure and concentration at each iterative step. The following equation is used for the pressure correction:

$$\nabla . (K . \nabla p') = \nabla . \vec{v}^* \tag{3.17}$$

Finally, the revised velocity is calculated from the following equation by using the corrected p'.

$$\vec{v} = \vec{v}^* - K \cdot \nabla p' \tag{3.18}$$

The convergence criteria are set to be equal to  $10^{-6}$ . Therefore, the relative error for all the unknowns (u,v,p,c,T) should be less than  $10^{-6}$ . Hence, the average relative error of the temperature, for example, can be calculated from the following equation:

$$\phi_F = \frac{1}{(n.m)} \sum_{i=1}^{i=m} \sum_{j=1}^{j=n} \left| (T_{i,j}^{k,s+1} - T_{i,j}^{k,s}) / T_{i,j}^{k,s+1} \right|$$
(3.19)

The iteration number showed by s and k marks the time step. The grid coordinates are denoted by i and j. In the in-house code where PC-SAFT EOS is used, the diffusion coefficients and the density are calculated at each point of the grid and are continually updated during the calculation procedure. The complete description of the algorithm can be found in [49, 50].

#### **3.3.1 Initial conditions**

The velocities at all grid points are initially set equal to zero. The initial concentration of water, either 0.9 or 0.5, is applied and the pressure of  $P_0=101325 Pa$  at the average temperature  $T_m = (T_{Cold} + T_{Hot})/2$  is calculated and used as an initial value. In each case, the time step  $\Delta t$  is obtained from the referred frequency by  $\Delta t = \prod/10$  where  $\prod = 1/f$  is the period of oscillation calculated from the associated frequency.

#### **3.3.2** Code validation

The results from FLUENT and the numerical in-house code were benchmarked against results from other research groups in two stages until good agreement was achieved. It will be described further in Chapter 4. In addition, the results obtained from the commercial software FLUENT was compared with the in-house code.

The analytical solution for the case without applied vibration, i.e. pure diffusion, was also compared to the numerical code results. In this regard, the separation of isopropanol between the hot and cold walls was extracted during the 12 hours until the system reached a steady state condition.

First the case with no vibration was considered. During the Soret separation phase, the concentration *C* satisfies:

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial y^2} \tag{3.20}$$

$$C(0, y) = C_0 (3.21)$$

$$\frac{\partial C}{\partial y}(t,0) = \frac{\partial C}{\partial y}(t,L) = \frac{\Delta C_{st}}{L}$$
(3.22)

Here  $C_0$  is the initial concentration, D is the diffusion coefficient, L is the diffusion path (cell size), and  $\Delta C = -S_T C_0 (1 - C_0) \Delta T$  is the steady state separation, where  $S_T$  is the Soret coefficient. The evolution of concentration in space and time is given by [41]:

$$C(t,y) = C_0 + \Delta C_{st} \left[ \frac{y}{L} - \frac{1}{2} + \frac{4}{\pi^2} \sum_{n,odd}^{\infty} \frac{1}{n^2} exp\left( -n^2 \frac{t}{t_D} \right) \cos\left(\frac{n\pi y}{L}\right) \right]$$
(3.23)

where  $t_D = \frac{L^2}{\pi^2 D}$  is the diffusion time. One can then easily determine the evolution of the concentration difference between the top and bottom of the experimental cell [41]:

$$\Delta C(t) = \Delta C_{st} \left[ 1 - \frac{8}{\pi^2} \sum_{n,odd}^{\infty} \frac{1}{n^2} exp\left( -n^2 \frac{t}{t_D} \right) \right]$$
(3.24)

where,  $\Delta C_{st}$  is the steady-state separation, which is the maximum possible separation. In this case, the maximum separation occurs as a result of the presence of the Soret effect and is calculated using the following equation:

$$\Delta C = -S_T C_0 (1 - C_0) \Delta T \tag{3.25}$$

In which time is denoted by t and  $t_D$  is the diffusion time that will be described further in the next section. The numerical analysis was performed for the similar case where the diffusion coefficients are set constant. The results in Figure 3.2 show complete agreement.



**Figure 3.2:** Comparison of numerical and analytical solutions for isopropanol separation between hot and cold walls.

#### **3.3.3** Mesh sensitivity analysis

Numerical errors are categorized as modeling, truncation, and computing errors. Modeling errors result when a complicated physical phenomenon is transformed into an idealized mathematical system. The mathematical system includes modeling parameters such as physical properties (diffusion coefficient, thermal conductivity, etc.) and initial conditions. The modeling parameters inevitably include uncertainties. Therefore, numerical results are also affected by these uncertainties. The effect of the modeling parameters on the numerical results can be estimated by a sensitivity analysis method.

Generally, since errors are unavoidable in numerical analyses, mesh sensitivity is necessary to check the accuracy of the results as well as reduce time and cost. In this study, I performed the sensitivity analysis through a two-dimensional diffusion problem.

Meshing is one of the most important and time consuming aspects of CFD modelling. Yet, steady advances in meshing technology and modern software tools make this task easier and more transparent. Meshes differ from mapped meshes to completely paved meshes to meshes with boundary layers to resolve gradients near the wall. A mapped mesh gives more control to the user whereas an automatic paver creates the mesh based on the algorithm employed. The choice of meshing technique depends on the problem and the physical properties at hand. The finer the mesh would be, the better the approximation. However, the density of the mesh also

directly correlates with the number of unknowns to be solved and has a direct impact on the computational effort to solve the problem.

The challenge is to know the type and size of the mesh that are appropriate to capture the details of the problem without exceeding the available time and computer resources. Furthermore, when solving coupled phenomena that have different dimensional scales, different mesh sizes at different locations of the domain are required. Ideally, the final solution should be independent of grid size, meaning the solution should not change if the mesh is further refined.

Although the numerical code was validated in the benchmark study, the grid convergence was checked for both FLUENT and the in-house code by using different mesh sizes in a mesh sensitivity analysis. Consequently, the most reasonable resolution was used for the analysis.

A systematic refinement of the mesh of at least two steps allows the formal definition of the error reduction. For the mesh sensitivity study, five meshes with different mesh refinements were generated as shown in Table 3.1. GAMBIT was used for generating the mesh and FLUENT was used for the analysis.

Mesh Type	No. of mesh Elements	The smallest element size (%L)
10x10	100	1
30x30	900	0.11
10x10 (f)*	100	0.3
20x20	400	0.25
20x20 (f)*	400	0.15

**Table 3.1:** Mesh resolutions used for mesh sensitivity analysis

\* f : finer mesh size near the boundaries.

In the model, water and isopropanol (0.5-0.5) were stored in a cubic cavity of 1 cm dimension in each side, and subjected to a temperature difference of  $10^{\circ}$ C in the top and bottom walls. Three different gravity magnitudes 0 (zero gravity),  $10^{-5}$  (micro gravity) and  $9.81 \times 10^{-2}$  (reduced gravity) were applied in the vertical direction (y). As the case is symmetric, a 2-D model was analyzed and simplified to a steady problem for the sensitivity analysis. For each case, the temperature and water mass fraction profiles along the middle vertical (AA in Figure 3.1) and

horizontal (BB in Figure 3.1) lines were recorded and analyzed. The obtained values were computed and compared. It was deduced from the related graphs (Figures 3.3 - 3.8) that the most sensitive physical parameter with which to optimize the mesh is the water concentration gradient for the case of  $g = 9.81 \times 10^{-2}$  as shown in Figure 3.8.



Figure 3.3: Temperature along the cavity between hot and cold walls at zero gravity



Figure 3.4: Water mass fraction along the cavity between hot and cold walls at zero gravity



Figure 3.5: Temperature along the cavity between hot and cold walls at micro gravity



Figure 3.6: Water mass fraction along the cavity between hot and cold walls at micro gravity



Figure 3.7: Temperature along the cavity between hot and cold walls at reduced gravity



Figure 3.8: Water mass fraction along the cavity between hot and cold walls at reduced gravity

It can be concluded from the above graph that the  $20 \times 20$  normal mesh is accurate enough for the actual analysis. It also shows that the finer mesh near the walls does not increase the accuracy of the analysis in this particular problem. The  $10 \times 10$  mesh size in both normal mesh and finer boundary mesh shows disharmony with the others. Finally, in order to have more accurate results,  $31 \times 31$  mesh size was used for the analysis.

In the in-house code, three meshes with different mesh refinements were generated. In the CFD model, the cavity was subjected to a vibration with frequency of 1 Hz and amplitude of 0.07m perpendicular to the imposed temperature gradient. The results were analyzed at two different characteristic times: viscous and thermal times. Velocity magnitude, isopropanol mass fraction and temperature along the middle vertical line (AA in Figure 3.1) were recorded and compared. The velocity magnitude and temperature profiles are shown in Figures 3.9 and 3.10. It can be concluded that the  $15 \times 15$  mesh size is accurate enough for the actual analysis. The  $10 \times 10$  mesh

size shows disharmony with the others. In general, for these particular problems a relatively course grid resolution should be sufficient as there is no strong flow in the cell.



Figure 3.9: Velocity magnitude along the cavity between hot and cold walls at zero gravity



Figure 3.10: Temperature along the cavity between hot and cold walls at zero gravity

# **Chapter 4**

# A Comparative Study of the Effect of Vibration on the Mixture at Different Conditions

## 4.1 Introduction

Water and isopropanol mixtures with two different compositions were selected for the first benchmark study. Three scientific groups were involved in the benchmark studies: Université Libre de. Bruxelles from Belgium, Perm University from Russia and our group at Ryerson University. The two mixtures were chosen in order to have both positive and negative Soret coefficients. The first one is a 50-50 mixture which has the positive Soret coefficient and the second is a 90-10 mixture that has the negative Soret coefficient as shown in Figure 1.1. The properties and specifications of the two studied cases are shown in Table 4.1. The values were obtained from the related ground based experiments. In order to simplify the problem, the fluid was modelled as a single fluid instead of a binary mixture. Therefore, the physical properties of the mixture (either 90-10 or 50-50) were used for modelling the fluid. Consequently, the effect of diffusion coefficients was not included in the analysis at this stage.

In the first benchmark, the effect of the vibration intensity was studied in the two different mixtures. The effect of the same level of vibration on the two different mixtures was studied by comparing the instantaneous velocity vector profiles and temperature contours. Also the line plots along the vertical and horizontal middle lines of the cavity were compared. In addition, the Nusselt number at the walls, average velocity and maximum velocity over the domain for the two cases using adiabatic and non adiabatic vertical walls compared and conclusion was made. In the second benchmark, the effect of different levels of vibration, both in terms of amplitude and frequency, was studied in the same two mixtures. Based on the experience achieved from the first benchmark, the mean velocity vector profiles over the domain and Nusselt number at the walls for all levels of applied vibration at different characteristic times for the two cases of adiabatic and non-adiabatic studied and the conclusion was made accordingly.

Viscosity [m <sup>2</sup> /s]	1.41E-06
Thermal diffusivity [m <sup>2</sup> /s]	1.30E-07
Diffusion coefficient [m <sup>2</sup> /s]	8.70E-10
Density [kg/m <sup>3</sup> ]	984
Thermal expansion [1/K]	3.10E-04
Solutal expansion	0.1386

Table 4.1: Physical properties of 90% - 10% (Top) and 50% - 50% (Bottom) mixtures

Viscosity [m <sup>2</sup> /s]	4.18E-06
Thermal diffusivity [m <sup>2</sup> /s]	8.50E-08
Diffusion coefficient [m <sup>2</sup> /s]	1.80E-10
Density [kg/m <sup>3</sup> ]	905
Thermal expansion [1/K]	7.70E-04
Solutal expansion	-0.25

# **4.2** Thermovibrational convection in the two mixtures under the influence of the same vibration

#### 4.2.1 Problem description and properties

As described, two mixtures were selected and the same vibration levels applied. The effect of vibration and temperature variation on the flow studied and compared between all scientific groups. These two cases are part of the IVIDIL experiment and the actual names used in the space experiments (Run#8 and Run#27) used herein. Modelling is performed ignoring diffusion, thus the liquid is assumed to be uniform. Calculations are done for two characteristic times. Initially the liquid has a mean uniform temperature and is motionless. The temperature of the liquid everywhere in the cell at t=0 s is equal to the mean temperature, i.e.  $T_{cold}+0.5\times\Delta T$ , where  $T_{cold}$  is the temperature at the cold wall and  $\Delta T$  is the temperature difference between the two walls, which will be imposed on the cell ( $T_{hot} - T_{cold}$ ). Boundary conditions are: rigid walls, either adiabatic lateral walls, or a linear temperature profile along the walls. The applied translation is having the form of X = A.  $Cos(\omega t)$  and is perpendicular to the temperature gradient as shown in Figure 3.1. The perpendicular angle ( $\pi/2$ ) shows the most effect between the applied vibration and the temperature gradient [34]. In summary, two tasks were assigned for all three groups as described below.

#### 4.2.2 Objectives specifications

Objective 1: Determine the dependence of the physical properties of the mixtures on time and concentration.

Objective 2: Calculation of the thermal convection assuming the vibration has the form x=A.cos(wt) and is perpendicular to the gravity vector for the water–isopropanol mixtures (two mixtures considered: 90% - 10% and 50% - 50%) with the following conditions:

- Vertical wall set to be adiabatic,
- ➢ Vertical wall set to have a linear temperature gradient.

The second objective was performed for a zero gravity (ideal condition) and for a static gravity condition reported from ISS (micro gravity, i.e.  $10^{-6}$ ). They were repeated for two cases: Run#8 (90% - 10%) and Run#27 (50% - 50%). The fluid was maintained at a temperature of 298K. Comparisons were carried out for the velocity profiles, flow pattern, and temperature distribution.

All three IVIDIL teams calculated the following:

- The profile of velocity at middle of the cell, i.e. Vx and Vz along the (x,y,z)=(x,0.5,0.5) and (0.5,0.5,z) lines,
- The flow structure at the vertical mid-plane y=0.5 after one viscous and two thermal times. More precisely, at 70.922 and 1538.44 seconds for Run#8 and 23.929 and 2352.94 seconds for Run#27.
- The times necessary for both velocity and temperature to reach steady state.

In the analysis, numerical simulations were carried out at different characteristic times. Viscous time (defined as  $t_{vs} = L^2/v$ ) is the time when the effect of vibration on the flow becomes noticeable. Thermal time (defined as  $t_{th} = L^2/\chi$ ) indicates when thermal equilibrium is established. Mass transport is significantly slower than the thermal process. Its characteristic time  $t_D = L^2/\pi^2 D$  (where D is the mass diffusion coefficient), being much larger than the thermal time (usually two orders of magnitude), is not used in the analysis.

#### 4.2.3 Results comparisons

The results were harmonised after a few iterations until complete agreement was achieved between the three groups. In the following graphs, velocity profiles ( $V_x$  and  $V_y$ ) and flow structures in Run#8 and Run#27 at one viscous and two thermal times compared for both adiabatic and non-adiabatic vertical walls. In the case of non-adiabatic vertical walls the temperature distribution is linear and is calculated using the following equation:

$$T_{\rm v} = 293 + 1000y \tag{4.1}$$

Here  $T_y$  is the temperature distribution along the vertical wall, 293 is the cold wall temperature, 1000 represents the  $\Delta T$  between the walls which is 10°K in Run#8 and Run#27 and y is the distance in millimetre changing from the cold wall along the vertical walls. The related User Define Function (UDF) used in FLUENT software is shown in Appendix A.



Figure 4.1: Flow structure in Run#8 at one viscous time, adiabatic vertical walls

Generally, the results compared qualitatively between the groups and the analysis repeated to obtain similar flow patterns. As shown in Figure 4.1, good agreement was achieved between the three groups after a few iterations. The slightly different flow structures are due to the different mesh sizes as well as numerical methods used for the analysis. While the full Navier-Stokes equations in the dimensional format was used by our group, other groups utilized the simplified equations in the non-dimensional form [40]. Figure 4.2 is the velocities along the X axis between

two adiabatic walls. The velocity values are very small in magnitude (i.e. in the order of  $10^{-6}$ ); therefore, the slight differences between them can be justified. Figure 4.3 shows the velocities along the Y axis, which is between the hot and cold walls. As mentioned earlier, due to the different mesh sizes used by the groups, small differences can be seen between the data sets. In all the graphs, the maximum velocity values as well as the velocity profiles are very close.



**Figure 4.2:** Velocity profiles  $(V_x \& V_y)$  along "*x*" axis in Run#8 at one viscous time, adiabatic vertical walls



**Figure 4.3:** Velocity profiles  $(V_x \& V_y)$  along "y" axis in Run#8 at one viscous time, adiabatic vertical walls

The flow structures at two thermal time are compared in Figure 4.4. No profile was presented by the Russian group, but the flow profiles are very similar between the other groups. However, flow direction is not the same due to the sinusoidal behaviour of the applied vibration. This is an acceptable difference as flow direction changes very quickly and slight differences in the recorded time can cause a large difference in flow direction. This can also be seen in the V<sub>y</sub> graph along the X axis and V<sub>x</sub> graph along the Y axis. The Belgium graphs are different from the

other groups in the direction and the maximum. The other two graphs show very close results for all three groups. As discussed before, this is due to different mesh sizes as well as the difference in equations used for the numerical calculations. Due to the small magnitude in the velocities these small differences are still in an acceptable range.



Figure 4.4: Flow structure in Run#8 at two thermal time, adiabatic vertical walls





Figure 4.5: Velocity profiles  $(V_x \& V_y)$  along "x" axis in Run#8 at two thermal time, adiabatic vertical walls





Figure 4.6: Velocity profiles  $(V_x \& V_y)$  along "y" axis in Run#8 at two thermal time, adiabatic vertical walls



Figure 4.7: Nusselt number on the cold wall after one and two thermal times in Run#8, adiabatic vertical walls [59]

The agreement between graphs was achieved after the fourth iteration in the calculations. As discussed earlier, they agree qualitatively and quantitatively. The Nusselt number was also calculated near the cold wall. The Nusselt number close to a value of one confirms the laminar flow behaviour near the boundaries, as expected. It also shows that convection and conduction are of almost similar magnitude with slightly more conduction heat transfer. It is slightly less than unity in the case of adiabatic walls. It oscillates with double frequency, i.e. 2 Hz, as shown in Figure 4.7.

The same analysis repeated for the case with non-adiabatic lateral walls with a linear change in the temperature as shown in Equation 4.1. The same agreement also achieved in this case at one viscous time in terms of both flow patterns as shown in Figure 4.8 and velocity line plots in the X and Y directions as shown in Figures 4.9 and 4.10. The flow directions are also the same and the plots show the same trend and maximum velocity magnitude for all the comparisons.



Figure 4.8: Flow structure in Run#8 at one viscous time, non-adiabatic vertical walls


**Figure 4.9:** Velocity profiles  $(V_x \& V_y)$  along "x" axis in Run#8 at one viscous time, nonadiabatic vertical walls



Figure 4.10: Velocity profiles  $(V_x \& V_y)$  along "y" axis in Run#8 at one viscous time, nonadiabatic vertical walls

At two thermal time, acceptable agreement was achieved between the groups both for the flow patterns as shown in Figure 4.11 and velocity line plots in Figures 4.12 and 4.13. The flow pattern obtained in our group is more similar to the Belgian one, where the Russians' flow pattern is slightly different particularly in the corners of the cell. Some differences in the flow direction can also be seen. The differences are greater between the groups in the two thermal

time in comparison with the one viscous time as expected, due to the longer time frame which results in the higher velocity magnitude in the Belgium results. This is acceptable as generally the velocity magnitudes are still in the range of  $10^{-4}$  m/s to  $10^{-6}$  m/s, which is very low.



Figure 4.11: Flow structure in Run#8 at two thermal time, non-adiabatic vertical walls





**Figure 4.12:** Velocity profiles  $(V_x \& V_y)$  along "x" axis in Run#8 at two thermal time, nonadiabatic vertical walls





**Figure 4.13:** Velocity profiles (V<sub>x</sub>&V<sub>y</sub>) along "y" axis in Run#8 at two thermal time, nonadiabatic vertical walls



Figure 4.14: Nusselt number on the cold wall after one and two thermal times in Run#8, nonadiabatic vertical walls [59]

Generally, the graphs agree in the case of non-adiabatic vertical walls. Similarly, the Nusselt number oscillates with double frequency, i.e. 2 Hz, as shown in Figure 4.14. It is slightly higher with non-adiabatic lateral walls in comparison with the adiabatic wall case. It is a bit more than one, which means that there is more convective heat transfer than conduction. Since it is still very close to unity, however, the flow is laminar as expected. It can also be concluded that thermovibrational convection is slightly stronger in the non-adiabatic cases in comparison with the adiabatic cases.



Figure 4.15: Nusselt number variation during the time on the cold wall in Run#8, adiabatic (top) and non-adiabatic (bottom) vertical walls [59]

Nusselt number variation during the whole time domain shows that temperature gets established faster when the vertical walls are non-adiabatic, i.e. there is a linear temperature profile along the wall, as shown in Equation 4.1. In the adiabatic case, the Nusselt number variation becomes steady at about 300 seconds whereas in the non adiabatic case, it is at about 200 seconds.



Figure 4.16: Max velocity variation during the time in Run#8, adiabatic (top) and non-adiabatic (bottom) vertical walls [59]

Velocity was also plotted during the first few viscous times (up to 160 seconds) in two different ways. The maximum velocity magnitude in the entire domain at each time step is shown in Figure 4.16. Average velocity was obtained at each time step by integration over the entire cavity. Both velocity graphs show that in the non-adiabatic case, velocity gets established sooner, similar to Nusselt number.



**Figure 4.17:** Average velocity variation during the time in Run#8, adiabatic (top) and nonadiabatic (bottom) vertical walls [59]

The same procedure repeated for Run#27 as shown for adiabatic lateral walls in Figures 4.18 to 4.23, both for the flow pattern and the velocity line plots. Generally, the flow pattern is more complicated in comparison with Run#8 due to the different composition weights, i.e. more isopropanol. Good agreement was achieved between the groups, but different flow directions were still seen due to the sinusoidal behaviour of the applied vibration. The magnitude of the maximum velocity was also within the acceptable range.



Figure 4.18: Flow structure in Run#27 at one viscous time, adiabatic vertical walls





**Figure 4.19:** Velocity profiles  $(V_x \& V_y)$  along "x" axis in Run#27 at one viscous time, adiabatic vertical walls





Figure 4.20: Velocity profiles  $(V_x \& V_y)$  along "y" axis in Run#27 at one viscous time, adiabatic vertical walls

The flow pattern was not provided by the Russian group for the two thermal time in the case with adiabatic walls. The result of the flow pattern obtained by our group is very close to Belgium's as shown in Figure 4.21. The velocity line plots also shows good agreement between the groups as shown in Figures 4.22 and 4.23.



Figure 4.21: Flow structure in Run#27 at two thermal time, adiabatic vertical walls



**Figure 4.22:** Velocity profiles  $(V_x \& V_y)$  along "x" axis in Run#27 at two thermal time, adiabatic vertical walls



**Figure 4.23:** Velocity profiles  $(V_x \& V_y)$  along "y" axis in Run#27 at two thermal time, adiabatic vertical walls



**Figure 4.24:** Nusselt number on the cold wall after one and two thermal times in Run#27, adiabatic vertical walls [59]

This agreement between all groups was achieved after several iterations in the calculations. The Nusselt number again oscillates with the double frequency of 2 Hz. For Run#27, its value is slightly higher than one, while for Run#8 its value was less than one. Therefore it shows the laminar behaviour of the flow as expected and the convection heat transfer is approximately equal to the conduction heat transfer near the boundary of the cold wall. As a result, thermovibrational convection is found to be slightly higher in the 50%-50% mixture in comparison with the 90%-10% mixture.

The same analysis was repeated for the non-adiabatic case and good agreement was achieved for the flow pattern as shown in Figure 4.25. The velocity line plots also follow the same trend between the groups and the maximum velocity values are in the acceptable range. The related graphs are shown in Figures 4.26 and 4.27.



Figure 4.25: Flow structure in Run#27 at one viscous time, non-adiabatic vertical walls





**Figure 4.26:** Velocity profiles  $(V_x \& V_y)$  along "x" axis in Run#27 at one viscous time, nonadiabatic vertical walls





Figure 4.27: Velocity profiles  $(V_x \& V_y)$  along "y" axis in Run#27 at one viscous time, nonadiabatic vertical walls

At two thermal time, the flow pattern in Figure 4.28 and velocity line plots in Figures 4.29 and 4.30, show complete agreement. The better agreement in two thermal time in this case and the other cases can be justified by the temperature distribution that becomes more stable after two thermal time in comparison with one viscous time.



Figure 4.28: Flow structure in Run#27 at two thermal time, non-adiabatic vertical walls



**Figure 4.29:** Velocity profiles  $(V_x \& V_y)$  along "x" axis in Run#27 at two thermal time, nonadiabatic vertical walls



**Figure 4.30:** Velocity profiles  $(V_x \& V_y)$  along "y" axis in Run#27 at two thermal time, nonadiabatic vertical walls



Figure 4.31: Nusselt number on the cold wall after one and two thermal times in Run#27, nonadiabatic vertical walls [59]

The Nusselt number in this case also oscillates at 2 Hz and its value was calculated near the cold wall and is shown in Figure 4.31. It is higher than in the adiabatic case of Run#27. This was similar in Run#8 when comparing the adiabatic case with the non-adiabatic. The same conclusion can also be made that there is more convective heat transfer near the boundary than conductive. Therefore, the thermovibrational heat transfer is greater in the non-adiabatic case of Run#27, which is a similar result to Run#8. The value close to unity still shows the laminar behaviour of the flow.



**Figure 4.32:** Nusselt number variation during the time on the cold wall in Run#27, adiabatic (top) and non-adiabatic (bottom) vertical walls [59]

The temperature also becomes stabilized more quickly in the non-adiabatic case for Run#27 as shown in Figure 4.32. It is about 300 seconds in the non-adiabatic case, but it is 450 seconds in the adiabatic case. Figures 4.33 and 4.34 also compare the velocity in the adiabatic and non-adiabatic cases of Run#27. Again having non-adiabatic lateral vertical walls make the velocities stabilize sooner than with adiabatic walls. This could also be due to the faster temperature stabilization in the non-adiabatic case.





**Figure 4.33:** Max velocity variation during the time in Run#27, adiabatic (top) and nonadiabatic (bottom) vertical walls [59]



**Figure 4.34:** Average velocity variation during the time in Run#27, adiabatic (top) and nonadiabatic (bottom) vertical walls [59]

#### 4.2.4 Conclusion

The benchmark results show that the establishing time in both temperature and velocity is shorter in the non-adiabatic vertical cases over the adiabatic cases. Thermovibrational convection is also slightly stronger in the case of non-adiabatic walls. In summary, three main conclusions can be made from the graphs:

- a. Generally thermovibrational convection is stronger in Run#27 with a 50%-50% mixture in comparison with Run#8 with a 90%-10% mixture, in both the adiabatic and non-adiabatic cases.
- b. The non-adiabatic cases, i.e. linear temperature distribution along the vertical wall, in both of the mixtures (50%-50% & 90%-10%) shows stronger thermovibrational convection as a result of the higher Nusselt numbers obtained.
- c. It can also be concluded from the following graphs that Run#8 with a 90%-10% mixture shows faster establishment in both temperature and velocity in comparison with Run#27 with a 50%-50% mixture. This was confirmed in both the adiabatic and the non-adiabatic cases. In other words, this behaviour can be justified by the stronger thermovibrational convection in the 50%-50% mixture in comparison with the 90%-10% mixture. The strong thermovibrational convection delays the establishment of temperature and velocity in Run#27.

In addition, it was concluded from the results that using micro gravity  $(10^{-6}g)$  or zero gravity for the analysis has similar outcomes. In spite of good agreements achieved after some iteration between the groups, it is difficult to analyze the instant snapshots of the flow because of change following the vibrations. Slight disagreements were noticed that could be attributed to somewhat different times used for making the plots. One group could take a flow picture at a slightly different moment than another and the results would be different. At the same time, the mean flow (either averaged in time over one period of oscillations in the case of full Navier-Stokes equations or simply calculated via the averaged approach) could be easily compared since it stays unchanged. The mean flow is the only result that does not contain the "slightly different time moment" problem of recording the data. Therefore, it was decided to use this approach for the second benchmark analysis that will be discussed in the next section.

# 4.3 Thermovibrational convection in two binary mixtures under various vibrations

### 4.3.1 Problem description

This benchmark study was focused on the identification of the vibrational convection intensity and enhancement of heat transfer. The results of this study will be used for the preparation and explanation of the experimental results. In vibrational convection, heat and mass transport is caused only by mean flow (called streaming in acoustic theory). In this study, eight cases called "Run" were performed with vibration applied at different levels of frequency and amplitude as shown in Table 4.3. The actual run numbers in the real experiment was selected herein. The frequencies, amplitudes and temperature differences were selected to have various ranges of Rayleigh vibration as shown in Table 4.3.

Runs	Water Mass Fraction	$\Delta \mathbf{T}$	Frequency (Hz)	Amplitude (mm)	Rav
Run 9	0.9	10	0.5	70	126.77
Run 10	0.9	10	0.2	70	20.28
Run 11	0.9	10	0.1	70	5.07
Run 12	0.9	10	0.05	70	1.27
Run 16	0.9	15	1	70	1140.96
Run 18	0.9	15	0.2	70	45.64
Run 19	0.9	15	0.1	70	11.41
Run 32	0.9	15	0.5	70	908.11

 Table 4.3: Runs and specifications

The vibrational analogue of Rayleigh number (Rayleigh vibration or Gershuni number) was used for defining the strength of the applied vibration:

$$Gs = Ra_{vib} = \frac{(A\omega\beta_T\Delta TL)^2}{2\nu\chi}$$
(4.2)

Where A is the amplitude of vibrations,  $\omega = 2\pi f$  is the angular frequency, L is the characteristic size,  $\Delta T$  is the applied temperature difference,  $\beta_T$  is the thermal expansion coefficient,  $\nu$  is the kinematic viscosity and  $\chi$  is the thermal diffusivity.

#### 4.3.2 Objectives

For each run two cases were studied:

- 1. Adiabatic case, Zero gravity
- 2. Non-adiabatic case (Linear temperature on the lateral walls), zero gravity

The boundary conditions and applied vibration were set similar to the previous benchmark as shown in Figure 3.1. The binary mixture without the Soret effect is considered. The initial state of the system used for the computation was no flow, mean uniform temperature  $(T_{hot}+T_{cold})/2$  and a uniform concentration.

The calculations were performed with time dependence until 2 thermal times, and the results were analyzed at four different characteristic times. The values are depending on the mixture composition and listed below:

- a) One viscous time: 70.92 s (90%-10%) or 23.93 s (50%-50%)
- b) Two viscous times: 141.84 s (90%-10%) or 47.86 s (50%-50%)
- c) One thermal time: 769.23 s (90%-10%) or 1176.47 s (50%-50%)
- d) Two thermal times: 1538.46 s (90%-10%) or 2352.94 s (50%-50%)

As described in the first benchmark, because of the difficulty in using snapshot analysis due to the change in the flow direction resulting from the sinusoidal aspect of the vibration, time-averaged analyses were used. The related mean velocities were obtained from the full Navier-Stokes equations and were calculated over 5 periods ( $\Pi$ =1/f):

$$\overline{T}(x, y, z, t) = \frac{1}{5\Pi} \int_0^{0+5\Pi} T(t) dt$$
(4.3)

$$\overline{u}(x, y, z, t) = \frac{1}{5\Pi} \int_0^{0+5\Pi} u(t) dt$$
(4.4)

where  $t_0$  is either one or two viscous and thermal times.

The following results were extracted for each run, at each characteristic time (a, b, c and d) for both adiabatic and non-adiabatic cases (A and B):

- I. The flow pattern: Vector velocity field and temperature isolines in the XY plane (Figure 3.1) when y = 0.5
- II. The mean velocity: Calculated over the volume from averaged velocity field determined by Equation 4.5:

$$u_{mean} = \frac{1}{Vol} \int \overline{u} (x, y, z, t) dx dy dz$$
(4.5)

III. Nusselt number: Nusselt number is the ratio of total convective and diffusive heat transfer to that in the purely diffusive regime (where Nu=1). For example, for horizontal walls the formula can be written as:

$$Nu_{cold,hot} = \frac{1}{L^2} \int_0^L \int_0^L \frac{\partial T}{\partial z} dx dy$$
(4.6)

The values in the cold and hot walls and lateral walls in both adiabatic and non-adiabatic cases were calculated and compared.

#### 4.3.3 Results comparisons

#### **4.3.3.1** Flow and temperature patterns

After several iterations, good agreements were achieved between the three groups. The results in micro gravity and zero gravity were almost identical so we preceded with the ideal zero gravity condition for all analyses. For the temperature and flow patterns, such a high number of graphs were produced that only the summary of the results is discussed here:

Firstly, in all the runs the four-vortex pattern was obtained in all of the characteristic times except sometimes in the initial one viscous time. This outcome will be discussed later in detail, but is expected based on the Gershuni theory [34]. Generally, the patterns of the mean flow are

due to the sinusoidal and translational behavior of the applied vibration. The comparison for Run#19, at two thermal time in the adiabatic case is shown in Figure 4.35.



Figure 4.35: Mean flow patterns in Run#19 at two thermal time for the adiabatic case

The temperature distribution was also always linear from the first characteristic time up to the final two thermal time. As an example, the temperature distribution is shown in Figure 4.36 for Run#12 at one viscous time in the adiabatic case. The temperature becomes established after a few seconds and therefore a linear distribution is always obtained in one viscous time. As a result, there was no difference between the non-adiabatic and the adiabatic cases. In both cases, the temperature becomes established before the first characteristic time.



Figure 4.36: Linear temperature distribution in Run#12 at one viscous time for adiabatic case

As mentioned earlier, there were some differences seen in the "mean flow pattern" at one viscous time. It is in the form of an unsymmetrical four-vortex pattern or even a one-diagonal pattern in some runs. It was found that this pattern is more pronounced in runs with a low Rayleigh vibration compared to those with a high Rayleigh vibration. For example in Figure 4.37, the flow pattern in Run#12 (the lowest Rayleigh vibration, i.e. 1.27) at one viscous time for the adiabatic case is shown. It is completely in the form of a one-diagonal pattern. It had changed, however, to the stable four-vortex pattern at two viscous times. This behavior was also found in the non-adiabatic case.



Figure 4.37: Mean flow pattern in Run#12 at one viscous time for the adiabatic case

In Figure 4.38, the mean flow pattern is shown for Run#11, which has the next lowest Rayleigh vibration (5.07) among all the runs in Table 4.3. The flow has the form of an unsymmetrical four-vortex pattern.



Figure 4.38: Mean flow pattern in Run#11 at one viscous time for the adiabatic case

Continuing the analysis to the runs with the next highest Rayleigh vibrations, the four-vortex pattern starts to appear at one viscous time. It can be concluded that by increasing the Rayleigh vibration, the thermovibrational convection increases and therefore the stable four-vortex pattern is obtained from the beginning. This behavior is also supported by both Nusselt numbers calculated near the hot and cold walls and by the average velocity magnitude in each run. They will be discussed further in the next part.

#### **4.3.3.2** Mean velocity and Nusselt numbers

The average velocity over the entire domain and Nusselt numbers at both the cold and hot walls were calculated for all of the runs and at all characteristic times. The summary of the results are discussed herein. There was good agreement achieved between the scientific groups as shown in Table 4.4 for the most critical run in terms of the applied Rayleigh vibration, i.e. Run#16. Our results are shown in red and compared with Belgium's that are in black.

	1 viscous time	2 viscous time	1 thermal time	2 thermal time
Average(V <sub>mean</sub> ) m/s	18.7700×10 <sup>-7</sup>	19.1930×10 <sup>-7</sup>	19.2000×10 <sup>-7</sup>	19.2160×10 <sup>-7</sup>
Average(V <sub>mean</sub> ) m/s	19.6044×10 <sup>-7</sup>	20.4191×10 <sup>-7</sup>	20.6696×10 <sup>-7</sup>	20.3385×10 <sup>-7</sup>
Nu <sub>cold</sub>	1.056905211	1.011002542	1.00989175	1.009956845
Nu <sub>hot</sub>	1.056766973	1.01077917	1.009696692	1.009708927
Nu <sub>cold</sub>	1.0532442	1.0013621-1.0014567	1.000045-0.0000693	1.000045-0.0000693
Nu <sub>hot</sub>	1.056905211	1.0013621-1.0014567	1.000045-0.0000693	1.000045-0.0000693

Table 4.4: The average velocity value and Nusselt numbers in Run#16 for the adiabatic case

The Nusselt numbers at each wall decrease over time, an effect which is due to the dynamics in the temperature field. It becomes stabilized within one thermal time. At the beginning the temperature is uniform and once different temperatures are applied at the walls and the vibration begins, a strong convection occurs, especially near the cold and hot walls where the largest density gradients reside due to thermal boundary layers. So, the Nusselt number strongly deviates from a value of one from the very beginning. Then, the velocity field becomes stabilized within one viscous time, and this effect mixes the liquid. This mixing plus diffusive process stabilizes the temperature over time. The Nusselt number changes with time and approaches a stable value, which is close to one in our case.

**Table 4.5:** Average velocity in all runs at four different characteristic times for both adiabatic and non-adiabatic cases

Runs	Run 12	Run 11	Run 19	Run 10	Run 18	Run 9	Run 32	Run 16
Ra <sub>v</sub>	1.27	5.07	11.41	20.28	45.64	126.77	908.11	1140.96
			I	Adiabatic			•	
1 viscous	4.039E-09	1.839E-08	2.799E-08	7.828E-08	1.262E-07	2.689E-07	7.645E-07	1.877E-06
2 viscous	4.949E-09	2.794E-08	4.215E-08	1.255E-07	1.943E-07	2.790E-07	7.807E-07	1.919E-06
1 thermal	4.801E-09	7.058E-09	1.210E-08	1.277E-07	1.977E-07	2.793E-07	7.821E-07	1.920E-06
2 thermal	4.062E-09	1.910E-08	2.911E-08	7.575E-08	1.238E-07	2.792E-07	7.825E-07	1.922E-06
	Non-Adiabatic							
1 viscous	4.694E-09	1.995E-08	3.025E-08	6.392E-08	1.283E-07	2.514E-07	7.705E-07	1.906E-06
2 viscous	4.961E-09	2.792E-08	4.208E-08	1.215E-07	1.939E-07	2.539E-07	7.782E-07	1.915E-06
1 thermal	4.801E-09	6.997E-09	1.195E-08	1.083E-07	1.975E-07	2.539E-07	7.784E-07	1.915E-06
2 thermal	4.061E-09	1.907E-08	2.903E-08	5.640E-08	1.235E-07	2.539E-07	7.788E-07	1.917E-06

The mean velocity values in all the runs with different levels of Rayleigh vibration are shown in Table 4.5 for all the characteristic times and in both the adiabatic and non-adiabatic cases.

The trend as shown in Figures 4.39 and 4.40 is for the average velocity that increase with an increase in the Rayleigh vibration, as expected.



Figure 4.39: Average velocity in the adiabatic case at all four characteristic times for all runs



Figure 4.40: Average velocity in the non-adiabatic case at all four characteristic times for all runs

The Nusselt numbers at the cold and the hot walls also increase with higher Rayleigh vibrations as shown in Tables 4.6 and 4.7. This can be explained by the increase in heat transfer which is due to the increase in the thermovibrational convection.

D //	Adiabatic		Non Adiabatic		D
Run #	one thermal	two thermal	one thermal	two thermal	Ka <sub>v</sub>
12	1.000323678	1.000164828	1.000189	1.0000793	1.27
11	1.000219636	1.000237989	1.0000488	1.0001222	5.07
19	1.000187229	1.000264572	1.000175	1.0001874	11.41
10	1.000409023	1.00037841	1.0002869	1.0002869	20.28
18	1.000528991	1.000492516	1.0006062	1.0005777	45.64
9	1.000476034	1.000463812	1.0014284	1.0013916	126.77
32	1.003129146	1.003239079	1.0032592	1.0033569	908.11
16	1.00989175	1.009956845	1.0118937	1.0118775	1140.96

Table 4.6: Nusselt number at the cold wall for all runs

Table 4.7: Nusselt number at the hot wall for all runs

Dup #	Adiabatic		Non Adiabatic		Pa
$\mathbf{R}$	one thermal	two thermal	one thermal	two thermal	Na <sub>V</sub>
12	0.999917088	0.999770287	0.9999291	0.9998681	1.27
11	0.999874157	0.999849674	0.9999658	0.9999047	5.07
19	1.000073653	0.999988097	1.0000777	1.0000858	11.41
10	0.999978027	0.999868142	1.0000634	1.0000572	20.28
18	1.000333997	1.000207752	1.0004724	1.0004316	45.64
9	1.00089341	1.000838602	1.0013269	1.0012963	126.77
32	1.003463015	1.003568714	1.0033978	1.0034996	908.11
16	1.009696692	1.009708927	1.011687	1.0117841	1140.96

The ascending trend of Nusselt numbers with increasing Rayleigh vibration in all the runs for both the adiabatic and the non-adiabatic cases are shown in Figures 4.41 and 4.42. This shows that by increasing the vibrational effect through either frequency and/or amplitude, the convective heat transfer is increased in comparison with the conductive heat transfer near the boundaries at the hot and cold walls. In other words, the thermovibrational convection heat transfer is increasing the Rayleigh vibration.

It can also be concluded that as time passes from one thermal time to two thermal time and further, the Nusselt number is increased. This shows that there is an increase in the convective heat transfer near the boundaries at the hot and cold walls. In addition, having non-adiabatic lateral walls intensifies the thermovibrational heat transfer near the boundaries, as expected. This can be verified by comparing the Nusselt numbers between the non-adiabatic and adiabatic cases of the same Runs.



Figure 4.41: Nusselt number variation at cold wall for all runs



Figure 4.42: Nusselt number variation at hot wall for all runs

#### 4.3.4 Conclusion

The second benchmark provided us more confidence in the use of our model, as well as some new ideas into the behaviour of flow due to vibration. After some iteration, complete agreement was achieved between all the groups. It was confirmed that the mean flow pattern is the most reliable way to compare the flow behaviour. As suggested by the theory [34], in all runs a fourvortex pattern was obtained, since the Rayleigh vibrations are all below the critical level [34]. The evolution of the flow was studied from one viscous time onward to the other characteristic times. It was concluded that in runs with a low vibration intensity, i.e. low Rayleigh vibration, the four-vortex pattern can appear later in comparison with runs with higher Rayleigh vibration. The Nusselt numbers at both the cold and hot walls and the average velocity over the entire domain were calculated for all runs in both the adiabatic and non-adiabatic cases at four different characteristic times. As expected, the Nusselt numbers increase from runs with low Rayleigh vibration to those with high Rayleigh vibrations. This can be used to explain the behaviour of the flows at one viscous time in runs with low Rayleigh vibration. Almost no significant difference was found between the adiabatic and non-adiabatic cases. However, in some runs, average velocities and Nusselt numbers were found to be higher in non-adiabatic cases compared to the same run with adiabatic vertical walls. This supports our conclusion in the first benchmark that thermovibrational convection is stronger in the non-adiabatic cases.

# **Chapter 5**

# The Soret Effect and Comparison Using Various Equation of States

## 5.1 Introduction

The analysis is continued by including the Soret effect in the model. The model is similar to the one shown in Figure 3.1, which consists of double diffusive convection in a binary mixture of isopropanol (10%) and water (90%) at zero gravity in the presence of the Soret effect. This will create a concentration gradient due to the temperature gradient between the walls. The direction of the displacement for each component of the mixture depends on the Soret coefficient sign. Based on studies by Pan *et al.* [21], for the current mixture, the Soret coefficient should be negative, which results in a high concentration of water in the hot side of the cavity.

Applying an external vibration to the cavity in a zero gravity environment makes the thermovibrational convection able to produce an increase in the heat transfer and a decrease in the concentration gradient. This is observable in a zero gravity condition due to the absence of buoyant convection. The component separation, or Soret separation, and the maximum temperature deviation from ideal conditions are the two parameters used to indicate the significance of the effect of convection induced by vibration on diffusion.

The vibrational analogue of the Rayleigh number, identified as Ra<sub>vib</sub> (Rayleigh vibration), was used for defining the strength of the applied vibration:

$$Ra_{\nu ib} = \frac{(A\omega\beta_T\Delta TL)^2}{2\nu\chi}$$
(5.1)

Where A is the amplitude of vibrations,  $\omega = 2\pi f$  is the angular frequency, L is the characteristic size,  $\Delta T$  is the applied temperature difference,  $\beta_T$  is the thermal expansion coefficient,  $\nu$  is the kinematic viscosity and  $\chi$  is the thermal diffusivity.

In this chapter, I examined the transport process (fluid flow, heat transfer and mass transfer) due to oscillatory vibration in the presence of Soret effect. The full transient Navier Stokes equations coupled with the mass and heat transfer formulation are solved numerically. The physical properties of the fluid mixture such as density are determined using two models, (i) the PCSAFT equation of state and (ii) the mass weighted mixing rule as shown in Table 5.1. The results of each model for the flow, temperature and concentration distributions are compared and analyzed in detail. Results show a significant effect of the selected density model on the flow pattern and components separation especially when subjected to vibrations with higher Rayleigh number.

The analysis was performed for three cases and in each case the mixing rule method (Equation 3.9) and PC-SAFT EOS [7, 8] were used for calculating the density. The diffusion coefficients were either calculated from PC-SAFT EOS or assumed constant in the mixing rule. Table 5.1 summarizes the two combinations of physical properties adopted for the analysis.

Properties	Comb.1	Comb.2	
Mass diffusion coefficient	D <sub>m</sub>	Constant	PC-SAFT EOS
Thermal diffusion coefficient	D <sub>T</sub>	Constant	PC-SAFT EOS
Density	ρ	Mixing rule	PC-SAFT EOS

**Table 5.1:** Combinations of physical properties

The related values for the 90% water and 10% isopropanol mixture calculated and used in the analysis are outlined in Table 5.2 below.

<b>Table 5.2:</b> 1	Density and	Diffusion	coefficients

Properties	Mixing rule	PC-SAFT EOS		
Density [kg/m <sup>3</sup> ]	$970.8\pm0.1$	$993.3\pm6$		
Mass Diffusion Coefficient [m <sup>2</sup> /s]	$7.77  imes 10^{-10}$	$7.77 \times 10^{-10} \pm 3.16 \times 10^{-11}$		
Thermal Diffusion Coefficient [m <sup>2</sup> /sK]	$2.39 \times 10^{-13}$	$2.39 \times 10^{-13} \pm 1.4 \times 10^{-14}$		
In addition, two different characteristic times were used for the comparisons: at one viscous time  $(t_{vs} = L^2/\nu)$  which is the time at which the flow becomes affected by the vibration, and at one thermal time  $(t_{th} = L^2/\chi)$  which is the time at which thermal equilibrium is established. Based on the density calculated in each model, the viscous and thermal times relative to each model are slightly different. This is due to the effect of density in the calculation of kinematic viscosity and thermal diffusivity as shown in Equations 5.2 and 5.3 respectively.

$$\nu = \mu/\rho \tag{5.2}$$

$$\chi = k/\rho C_p \tag{5.3}$$

In the following graphs, the first value for the characteristic time is always calculated from the mixing rule method and the second time is from the PC-SAFT EOS method. In Table 5.3 the characteristic times for each method is shown.

Characteristic times [s]	Mixing rule	PC-SAFT EOS
Viscous time	93.7	95.8
Thermal time	742.2	759.4
Diffusion time	$129 \times 10^3$	$129 \times 10^3$

Table 5.3: Characteristic times

In the first case, as shown in Table 5.4, the temperatures are set at 293°K and 303°K for the cold and hot walls. The applied frequency and amplitude are 0.05Hz and 0.07m respectively. As a result, based on the densities calculated from the related model, the Rayleigh vibrations are 1.62 and 1.7 for the mixing rule method and PC-SAFT EOS, respectively. In the second case, the temperatures and amplitude remain unchanged but the frequency increased to 1Hz in order to achieve Rayleigh vibrations equal to 646 and 679 respectively. The third case has the hot wall temperature set equal to 308°K, the frequency increased again to 2Hz and the amplitude is set to 0.057m. As a result, the Rayleigh vibrations are found to be 3857 and 4038 for the two studied models, respectively.

Properties	Case 1	Case 2	Case 3	
Cold wall temperature [K]	293	293	293	
Hot wall temperature [K]	303	303	308	
G-jitter Frequancy [Hz]	0.05	1	2	
G-jitter Amplitude [mm]	70	70	57	
Rayleigh Vibration	1.62/1.7 1	646/679 <sup>1</sup>	3857/4038 <sup>1</sup>	
<sup>1</sup> Based on the density calculated from mixing rule method and PC-SAFT EOS				

Table 5.4: Three studied cases with different applied conditions

The temperatures, frequencies, and amplitudes were selected in order to have different levels of Rayleigh vibration as shown in Figure 5.1. The value 8000 is the critical value suggested by the theory [34] for flow evaluation.



**Figure 5.1:** Distribution of Rayleigh vibration (Mixing rule: ▲ PC-SAFT EOS ♦ )

# 5.2 Case 1: Low Rayleigh vibration $(Ra_{vib} = 1.62 / 1.7)$

The temperature difference is set 10°K and a very low frequency of 0.05Hz is applied. The applied amplitude is also 0.07m. As a result, the Rayleigh vibrations are 1.62 and 1.7 for the two models. The temperature and isopropanol mass fraction in the middle vertical line (Line AA in Figure 3.1) are plotted at one thermal time as shown in Figure 5.2.



Figure 5.2: Temperature and isopropanol mass fraction at one thermal time (t=742 s and 752 s)

The results show complete agreement between the two density models. When thermal diffusion forces the two components to separate between the hot and cold walls, the applied external vibration makes the components mix. The flow created by the imposed vibration was weak enough that it had no effect on the temperature distribution in the cavity. As such, a linear temperature distribution was obtained regardless of the way the density is evaluated.

The instantaneous velocities at each node were averaged during the five periods after each characteristic time for better comparison. The mean velocity vector profiles for PC-SAFT EOS at one viscous and one thermal time are shown in Figure 5.3.



Figure 5.3: Mean velocity vector profile at one viscous time, t=96 s and at one thermal time, t=752 s using PC-SAFT EOS

Based on the Gershuni theory [34] the four-cell pattern was detected regardless of the time selected. Figure 5.3(b) shows the four-cell flow pattern using PC-SAFT EOS, which is in agreement with Gershuni theory.

In order to study the instantaneous variation of the parameters in the domain, two critical points in the domain, P1 and P2, as shown in Figure 3.1, were adopted. The rate of change in density is plotted at point P1 for the two models as shown in Figure 5.4. The values are about 10<sup>3</sup> times higher using PC-SAFT EOS when compared with the mixing rule method. As expected, the rate of change of density was higher at the start but decreased significantly after one viscous time. Using the mixing rule to calculate the density showed no change in the density after one viscous time, whereas for the density calculated using PC-SAFT EOS a much higher rate of change during the whole process was found.



Figure 5.4: Rate of change of density at point P1 using mixing rule method and PC-SAFT EOS

This behaviour makes the average velocity in the domain for PC-SAFT EOS much higher in comparison with the mixing rule method as shown in Figures 5.5 and 5.6. There is a  $10^4$  order of magnitude difference between the velocities in the two models, with the higher velocity calculated when PC-SAFT EOS is used.



Figure 5.5: Average velocity using the mixing rule method and PC-SAFT EOS



Figure 5.6: Average velocity using the mixing rule method and PC-SAFT EOS



Figure 5.7: Density and temperature variation at point P1 using the mixing rule method



Figure 5.8: Density and temperature variation at point P1 using the PC-SAFT EOS

This behaviour can be due to the strong link between instantaneous variations of density and temperature in the PC-SAFT EOS, which is not found in the mixing rule method, as shown in Figures 5.7 and 5.8. Instantaneous variation in the temperature makes the density fluctuate much more using PC-SAFT EOS compared to the mixing rule approach.

## 5.3 Case 2: Intermediate Rayleigh vibration ( $Ra_{vib} = 646 / 679$ )

In order to get higher Rayleigh vibration, the frequency was increased to 1 Hz. The temperature and amplitude were set the same as in the previous case. As a result, the Rayleigh vibration increased to about 646 and 679 based on the adopted density model. The temperature and isopropanol mass fraction line plots at one thermal time are shown in Figure 5.9.



Figure 5.9: Temperature and isopropanol mass fraction at one thermal time (t=742 s or 752 s)

The temperature profile was linear when the mixing rule method was used at both characteristic times, whereas the PC-SAFT EOS at the same times showed some nonlinearity. This means that the temperature is influenced by the external vibration when the density is calculated by PC-SAFT EOS. Concentration plots for the isopropanol mass fraction also show disagreement

between the two models. The mixing rule method presents more component separation in comparison with the PC-SAFT EOS. This difference is higher at one thermal time than at one viscous time, which indicates an increase in the disparity between the two models by that time.



Figure 5.10: Mean velocity vector profile at one thermal time using the mixing rule method (unsymmetrical four cell pattern)

The mean velocity vector profile during the 5 periods for the mixing rule method did not predict the complete four-cell pattern at one viscous time and showed unsymmetrical shape of the four cells at one thermal time, as shown in Figure 5.10. In contrast, PC-SAFT EOS predicted the full symmetrical configuration of the four cell pattern at both one viscous and one thermal times.

The rate of change in density at point P1 for the two models is shown in Figures 5.11 and 5.12. The rate of change is about  $10^3$  times higher in PC-SAFT EOS compared to the mixing rule method. Similar to the previous case, the rate of change of density is higher at the beginning, but decreased significantly after one viscous time. The mixing rule method showed much less fluctuation in the density whereas PC-SAFT EOS had a much higher rate of change of density.



Figure 5.11: Rate of change of density at point P1 using the mixing rule method and PC-SAFT EOS



Figure 5.12: Rate of change of density at point P1 using the mixing rule method and PC-SAFT EOS



Figure 5.13: Average velocity using the mixing rule method and PC-SAFT EOS

The disagreements between the mean velocity vector profiles can be explained by the average velocity magnitude differences between the two models, as shown in Figure 5.13. There are about  $10^4$  order of magnitude differences between the average velocities in the two models. The higher velocity magnitude in PC-SAFT EOS causes more powerful mixing that result in less component separation in comparison with the mixing rule method as shown in Figure 5.9.

This behaviour can also be explained by the strong link between density and temperature in the PC-SAFT EOS, which is not found in the mixing rule method. The density varied much more in PC-SAFT EOS in comparison with the mixing rule method. In order to study the instantaneous variation of these parameters, the same point P1 was examined. Figure 5.14 shows the density and temperature variation at this point up to two thermal times in the mixing rule method.



Figure 5.14: Density and temperature variation at point P1 using the mixing rule method



Figure 5.15: Density and temperature fluctuations using the mixing rule method

The variation in the density is very low and does not show a link with the temperature. When the temperature became steady at about one viscous time, density continued its descending trend. The fluctuations were also shown in Figure 5.15 during 5 periods around one thermal time, which showed no link between temperature and density in the mixing rule method.

In the PC-SAFT model, the behaviour is completely different as shown in Figures 5.16 and 5.17. Density and temperature are coupled and have more instantaneous fluctuations in comparison with the mixing rule method.



Figure 5.16: Density and temperature variation at point P1 using PC-SAFT EOS



Figure 5.17: Density and temperature fluctuations during 5 periods around one thermal time using PC-SAFT EOS

# 5.4 Case 3: High Rayleigh vibration ( $Ra_{vib} = 3857 / 4038$ )

In the third case, the Rayleigh vibration was set to be around 4000. The temperature and concentration plots in Figure 5.18 show high amounts of disagreement between the two models. The temperature distribution in PC-SAFT EOS was distorted when it was still linear in the mixing rule method.

The separation of components at one thermal time was more pronounced using the mixing rule method as shown in Figure 5.18. This is in contrast to the strong mixing using the PC-SAFT EOS.



Figure 5.18: Temperature and isopropanol mass fraction at one thermal time (t=742 s or 752 s)



Figure 5.19: Mean velocity vector profile (One diagonal vortex pattern) at one thermal time using PC-SAFT EOS

The four-cell pattern was expected in this case as the Rayleigh vibration is below the critical value  $Ra_{cr}$ =8000 [34]. Based on the work of Gershuni *et al.* [34], above the critical value the four-cell pattern should change to a one–diagonal pattern, and the temperature profile should be distorted. In PC-SAFT EOS, although the Rayleigh vibration is far below the critical value, the temperature profile unexpectedly distorted and the one-diagonal vortex pattern was obtained at one thermal time, as shown in Figure 5.19. In the mixing rule method, as expected, the four-cell pattern was observed at one thermal time.



Figure 5.20: Rate of change of density at point P1 using the mixing rule method and PC-SAFT EOS

The rate of change in density is plotted at point P1 for the two models as shown in Figures 5.20 and 5.21. The rate of change is about  $10^4$  times higher using PC-SAFT EOS compared to use of the mixing rule method. The higher velocity magnitude in PC-SAFT EOS makes powerful mixing and therefore less component separation than with the mixing rule method.

In Figure 5.22, the magnitude of the average velocity in the domain was plotted during 5 periods near one thermal time for the mixing rule method and PC-SAFT EOS. As shown, the average velocity was much higher when using PC-SAFT EOS rather than the mixing rule method at both characteristic times.



Figure 5.21: Rate of change of density using the mixing rule method and PC-SAFT EOS



Figure 5.22: Average velocity during 5 periods around one thermal time using the mixing rule method and PC-SAFT EOS

While the velocity from the mixing rule method fluctuated in the range of E-10 and E-12, it varied in a range of E-6 and E-7 when using PC-SAFT EOS. The situation was similar at one viscous time. There was about  $10^4$  order of magnitude difference between the velocities in the two methods.

This behaviour can be explained through temperature and density variations. Figure 5.23 shows the density and temperature variation at the critical point P1 (described above) up to two thermal times in the mixing rule method.



Figure 5.23: Density and temperature variation at point P1 using the mixing rule method

The variation in the density is very small and does not show a link with the temperature. The temperature profile became steady at about one viscous time, but density continued its descending trend. The instantaneous fluctuation is shown in Figure 5.24 during the 5 periods around one thermal time. It also shows no link between the temperature and density using the mixing rule method.



Figure 5.24: Density and temperature fluctuations during 5 periods at about one thermal time using the mixing rule method

Using PC-SAFT EOS, as shown in Figure 5.25, the density and temperature are strongly coupled. Unlike the two previous cases, there are two non-uniformities in the temperature and density curves before they become steady. This happens around one thermal time and more specifically between 400 to 800 seconds, as shown in Figure 5.25. This behaviour can be described by an increase in the velocity magnitude that reaches a specific stage. Accordingly, since the velocities are strong enough, they can influence the composition and temperature isolines. This is due to the higher Rayleigh vibration in this case in comparison with the other two cases. Correspondingly, the transmission between four cell pattern and one diagonal which happens also between 400 and 800 seconds can be explained by considering that the PC-SAFT EOS combination has higher instantaneous fluctuations in comparison with the mixing rule method as shown clearly in Figure 5.26 in the 5 periods around one thermal time.



Figure 5.25: Density and temperature variation at point P1 using PC-SAFT EOS



Figure 5.26: Density and temperature fluctuations using PC-SAFT EOS

The disagreements between the velocity vector profiles along with the velocity magnitude differences between the two combinations are the reasons for the disparities in the temperature and concentration plots in Figure 5.18. The higher velocity magnitude in PC-SAFT EOS makes the mixing much more powerful, which results in less component separation in comparison with the mixing rule as shown in Figure 5.18. Also, in PC-SAFT EOS the temperature profile is highly influenced by the velocity profile due to its magnitude and shape.



Figure 5.27: Temperature and mean flow patterns using the mixing rule  $(\Delta T \text{ Increments} \approx 0.8 \text{ }^{\circ}\text{C})$ 



Figure 5.28: Temperature and mean flow patterns using PC-SAFT EOS ( $\Delta T$  Increments  $\approx 0.8$  °C)

Due to the discussed differences between the rate of change of density, the mean velocity vector profiles and the average velocity magnitudes, the temperature and concentration patterns and plots are quite different in this case than for the two studied models as shown in Figures 5.27 and 5.28.

The thermal diffusion coefficient variation is also compared in this case by using PC-SAFT EOS with an ideal no-vibration case ( $Ra_v = 0$ ). As expected, the distribution of the thermal diffusion coefficient is linear when Rayleigh vibration is set equal to zero. The pattern is gradually distorted when Rayleigh vibration was increased. Figure 5.29 shows the comparison between the no vibration case with this high vibration case, i.e.  $Ra_v = 4038$ . It clearly shows the importance of having variable thermal diffusion coefficients when using this equation of state, especially in cases with high Rayleigh vibration.



**Figure 5.29:** Thermal Diffusion Coefficient variation at Gs = 0 (—) and Gs = 4038(---)

# 5.5 Conclusion

Double-diffusive thermal convection with the Soret effect is considered in a cubic cell subjected to external vibrations. The structure of the time-averaged (mean) fields was examined on a long time scale, i.e. during thermal time, along with the composition and temperature profiles. Three different levels of Rayleigh vibrations were considered by using different frequencies, amplitudes and temperature gradients. Two models, weighted average mixing rule and PC-SAFT EOS were used for calculating density. It was found that using PC-SAFT EOS makes the formation of a strong one-diagonal flow pattern in Rayleigh vibrations less than the critical number Gershuni theory predicted. Accordingly, significant mixing occurs when using PC-SAFT EOS that results in a decrease of component migration. The velocity magnitude is higher when the density calculated using PC-SAFT EOS makes higher instantaneous changes in the density. This results in higher velocity magnitudes which then produce less component separations.

# **Chapter 6**

# **Experimental and Numerical Results Comparison**

## 6.1 Introduction

The experiment IVIDIL (Influence of Vibrations on Diffusion in Liquids) was performed in fall 2009 onboard the ISS, inside the SODI (Selectable Optical Diagnostics Instrument) mounted in the glove box on the ESA Columbus module. It was intended to be used to carry out 41 experimental runs with each of them lasting 18 hours. The objectives of the experiment are outlined below:

- After each space experiment, there was a discussion about the role of onboard g-jitters. The experiment helped to identify the limit level of vibrations where g-jitter does not play a role for onboard experiments. This was satisfied by observing the diffusive process under different imposed and controlled vibrations.
- 2. To perform accurate measurements of diffusion and thermal diffusion coefficients for two binary mixtures in the absence of buoyant convection. The measured values can be used as standards for ground experiments. Two different concentrations of waterisopropanol (IPA) with positive and negative Soret effect were used as test fluids. This objective is also related to the influence of vibrations on the measured values of diffusion and thermal diffusion coefficients.
- 3. Finally, to investigate vibration-induced convection and, particularly, heat and mass transfer under vibrations.

Three international teams were involved in the project. Other than our team at Ryerson University led by Dr. Saghir, a Team from Université libre de Bruxelles was responsible for aspects related to IVIDIL experimental definition, theoretical and numerical modelling, and a Russian team from Perm University provide theoretical and numerical support.

ESA's SODI multi-user facility (Selectable Optical Diagnostics Instrument) was initially developed for conducting three experiments: IVIDIL, DSC and Colloid. It was installed on September 23, 2009. Hardware development was provided by Verhaert Space NV (Kruibeke, Belgium) and optical development was provided by Lambda-X (Nivelles, Belgium).

The first experiment of IVIDIL, started on October 5, 2009 and a total of 55 experimental runs (41 original runs and 14 repetitions) were successfully completed by January 20, 2010. Each original run lasted 18 hours and all of them were controlled via tele-science provided by the Spanish User Support Center (E-USOC, Madrid).

All experiments conducted inside the SODI facility use available optical diagnostics. The IVIDIL experiment employs Optical Digital Interferometry and Particle Image Velocimetry. The IVIDIL instrument consists of three principal parts (see Figure 6.3):

(1) Mach–Zehnder Interferometer joining with the equipment for digital recording of the phase information;

- (2) The diffusion cell;
- (3) Vibrational stimuli.

The prototypes of all parts were designed, developed and tested on the ground [41] or during experiments performed in parabolic flights [25, 42].

Although g-jitter seems to have a major impact on diffusion-related experiments in Space, very few experimental studies have addressed this topic. One of the first experiments studying the impact of residual accelerations on convection onboard a spacecraft in differentially heated cylindrical cavities was performed by Naumann et al. [51] and independently by Babushkin et al. [52]. In these experiments, the temperature was monitored at several fixed points. The results did not provide clear evidence of time-averaged flows nor of the related heat transfer caused by periodic high-frequency g-jitter.

MEPHISTO (from the French descriptive name "Material pour l'Etude des Phenomenes Interessant la Solidification sur Terre et en Orbite") was a series of cooperative investigations between NASA, the French Space Agency (CNES), and the French Atomic Energy Commission (CEA). The French Space Agency developed the MEPHISTO experiment apparatus and NASA provided the flight opportunities on the space shuttle. The MEPHISTO experiments studied the role of gravity-driven convection during the solidification process in materials. The MEPHISTO apparatus was a complex directional solidification furnace that provided information on the solidification in real-time. Of particular interest was the See-beck thermoelectric signal that measured the temperature and thus, through the phase diagram, the composition of the solid-liquid interface. The results achieved from the MEPHISTO experiments were ultimately used to improve materials preparation and processing on Earth.

The MEPHISTO data from the USMP-3 Space flight mission in 1996 by Garandet et al. [53] represented the first quantitative data on g-jitter effects in a directional solidification experiment. However, it was concluded that there is a lack of well-documented experimental results that can be used for verification of numerical modelling to provide reliable predictions of g-jitter effects.

In addition, some experiments were carried out by Smith et al. [38] onboard the Mir station using the Canadian MIM (Microgravity Isolation Mount) facility. The MIM provides the opportunity for exposing the diffusion couples to the ambient g-jitter of Mir, isolating them from this and also subjecting them to a forcing vibration superimposed on the isolating state. Their primary analysis indicated that diffusion coefficients in dilute binary metallic alloys depend upon the residual accelerations and the quality of microgravity. Joint Russian–German experiments onboard Foton M3 in 2007 was directed towards studying the influence of g-jitter on solidification.

Thus, there has consistently been a lack of experimental data quantifying onboard g-jitter effects, which demonstrates the necessity of such a Space experiment. In this chapter, I was analyzed the experimental results in the form of laser images using image processing tools. The experimental results for each specific run (Refer to the specific cases called "Run" with specific applied frequency and amplitude) analyzed to plot the concentration gradient between the walls during the 18 hours experiment duration. Also, the same scenario modelled using both Commercial software FLUENT and in-house code using PC-SAFT EOS. The concentration gradient during the time plotted and compared for all the runs with various Rayleigh vibrations i.e. various

frequency and amplitude. The comparison was performed and the conclusion was made in comparing the numerical and experimental results at each specific run and also between different runs i.e. different levels of vibration.

#### 6.2 Experiment and the pre-flight preparations

As discussed in the previous chapters, the presence of convective flows in diffusion cells will significantly alter the concentration fields. For this reason, residual convection has to be reduced as much as possible. When the thermal gradient is directed against gravity, the mechanical equilibrium in the cell is generally stable. However, convection can easily appear at the lateral walls of the cell and in the corners where thermal perturbations are unavoidable. The effect of convection is more pronounced in the case of a negative Soret coefficient, when the heavy liquid goes to the hot wall (against gravity). Therefore, performing diffusion controlled experiments in space where gravity is absent is a unique method to remove the effect of residual convection from the measurements.

Before the actual experiment on board the ISS, some related experiments were performed in the ground-based lab. The ground-based experiments in IVIDIL included the study of two physical phenomena with very different time scales: slow diffusion and fast vibrations. Both of them are affected by buoyant convection on the ground. However, some features of vibrational convection can be analyzed during short microgravity time (22s–25s) in parabolic flights. The experimental evidence of averaged flows in non-uniformly heated fluids under vibrations in a microgravity environment has been confirmed in parabolic flight experiments [25, 42]. Numerical simulations before the flight are able to combine both phenomena and make predictions of expected experimental development. Therefore, as discussed in the previous sections, numerical simulations performed before the actual experiments.

### **6.3** Experiment principals

The numerical models are based on different assumptions and thus not a complete picture of the phenomena. As a result, they should be more extensively verified by experimental measurements. A variety of different experimental techniques were used for the measurements of

Soret effect in liquids. The experimental methods can be roughly divided into two classes: (1) methods that employ the presence of buoyant convection in the measurements; (2) methods in which buoyant convection plays an adverse role and efforts are done to minimize its effect.

The measurement method used in the second group is optical method which relies on the variation of refractive index with composition. The main features of optical methods are non-intrusiveness and sensitivity. The main advantage of these methods is the absence of mechanically driven parts in contact with liquid, so measurements do not disturb the experiment. The main disadvantage is that the media under investigation must be transparent. Recent advances in lasers and electronic cameras as well as the increasing processing power of computer-aided data processing have enabled considerable progress in the development of new optical measurement techniques, along with a revival of traditional techniques.

## 6.4 Strategy of the experiment

The initial idea of the IVIDIL experiment was to measure diffusion coefficients in binary mixtures under different vibrational frequencies and amplitudes. To do this, concentration non-uniformity in the liquid is required. If the mixture is prepared on Earth, the components would diffuse during the waiting time on the launch site. Therefore, by applying a temperature gradient, thermal diffusion (the Soret effect) can be used to produce a concentration gradient in an originally uniform mixture. Thus, the Soret effect will be employed for creating a concentration profile across the cell. As a result, the objectives of the experiment were extended to include the measurements of the Soret coefficient as well, as it has industrially and scientifically driven interests.

Accordingly, each experimental run is performed in two steps. During the first step (duration of 12 hours) a temperature difference  $\Delta T$  is imposed between horizontal walls of the experimental cubic cell. Due to the Soret effect, the concentration gradient is slowly generated in an initially homogeneous binary mixture. In the ideal condition, when other disturbances like vibrations, and convection are absent, the time evolution of the concentration difference between hot and cold walls within the cell  $\Delta C(t) = C_{hot}(t) - C_{cold}(t)$  can be described by [41]:

$$\Delta C(t) = -S_T C_0 (1 - C_0) \Delta T \left[ 1 - \frac{8}{\pi^2} \sum_{n,odd}^{\infty} \frac{1}{n^2} \exp\left(-n^2 \frac{t}{\tau_r}\right) \right]$$
(6.1)

where  $S_T$  is the Soret coefficient,  $\tau_r = L^2/\pi^2 D$  is the relaxation time, L = 0.01 m is the cell size, D is the diffusion coefficient,  $\Delta T$  is the applied temperature difference, and C<sub>0</sub> is the initial concentration of the mixture. This stage is shown in Figure 6.1.

By using  $\Delta C(t)$  from the experiments, we can determine  $S_T$  and D from Equation 6.1 through a mathematical fitting procedure. Obviously, in the presence of vibrations,  $\Delta C(t)$  will not agree completely with the theoretical curve generated by Equation 6.1. Therefore, the other major output of the IVIDIL experiment is the set of curves of  $\Delta C(t)$  for different levels of vibrations. Consequently, effective coefficients of diffusion and thermal diffusion can be determined. From the magnitude of the difference between the effective and theoretical D and  $S_T$  coefficients, one can determine the vibrational level at which a process can be considered a diffusion controlled phenomena.

In the second step (duration of 6 hours), the system is reverted to an isothermal case where the temperature is kept constant at  $T_{mean} = 25$  °C. The characteristic thermal time is relatively short  $\tau_{th} = L^2/\chi \approx 12min$  ( $\chi$  is thermal diffusivity). Note that the characteristic diffusion time is much larger,  $\tau_D = L^2/D \approx 32h$ . During this step, diffusion will progressively reduce the previously established concentration gradient as depicted in Figure 6.1. The diffusion coefficient D can also be determined from this step.

An additional advantage of this experimental technique is that it provides the opportunity to track backward diffusion relaxation after switching off the temperature gradient applied to the cell. The measurement of coefficients in two steps allows a further increase in accuracy, as the diffusion coefficient is measured twice. Then, the corrected diffusion coefficient can be introduced into equations for thermal diffusion to improve the accuracy of the Soret coefficient fit.

Generally, the choice of experiment duration is a compromise between relaxation time and available microgravity time. Diffusion relaxation time for the 90% water -10% isopropanol

mixture is  $\tau_r = L^2/\pi^2 D \approx 3.25h$ , and the 12 h duration of the non-isothermal step is sufficient to approach steady state. Here again *L* is the cell size in the direction of temperature gradient and *D* is the mass diffusion coefficient.

As mentioned, the procedure shown in Figure 6.1 was repeated 41 times under vibrations of different frequencies and amplitudes, or without them to measure the natural level of g-jitters onboard the ISS. The evolution of the concentration distribution during the two steps was tracked by recording digital interferograms.



**Figure 6.1:** Numerical simulation of a typical IVIDIL experiment at the absence of vibrations. Component separation with time for liquid with negative Soret (90% water – 10% isopropanol)

### 6.5 Experimental scenario

Generally, the experiment was performed for each run as follows:

- 1. 25 min for establishment of the mean temperature
- 2. 5 min to establish the temperature gradient.

Hot wall:  $T=25^{\circ}C + \Delta T/2$ ; Cold wall  $T=25^{\circ}C - \Delta T/2$ 

- 3. Vibrations are switched on for 18 hours
- 4. The temperature gradient switched off after 12 hours
- 5. 6 hours for isothermal diffusion with the vibration switched on

The experiments were performed in a transparent cubic cell filled initially by a homogenous mixture. The thermal gradient is imposed by heating and cooling the top and bottom walls of the cell, respectively. The spatial temperature variation produces mass transfer through the Soret effect. An enlarged collimated laser beam traverses the entire cell perpendicular to the temperature gradient. Both temperature and composition variations contribute to the refractive index. Using an interferometric technique the final wave-front shape is accurately determined by comparison with the undisturbed shape. The optical interferometer coupled with a digital recording and processing unit is used for very accurate determination of phase shift. Each interferogram is reconstructed by performing a 2D Fast Fourier Transform (FFT) of the fringe image, filtering a selected band of the spectrum, performing an inverse 2D FFT of the filtered result and phase unwrapping. The details of this procedure will be described further in the next part. Knowledge of phase shifts gives information about the local gradients of the refractive index. Then, the gradients of composition inside of the fluid are calculated based on the refractive index gradients and local temperatures. Thus, optical digital interferometry enabled measurements of  $\Delta C$  between two arbitrary points which are near the hot and cold walls in the cell.

The set-up was developed using the concept of a Mach – Zehnder interferometer. As discussed before, in the experiment, an optical cubic cell of internal size L = 10 mm was used, which was made of quartz Suprasil by the Hellma company. The top and bottom of the cell are kept at constant temperatures (T<sub>hot</sub> and T<sub>cold</sub>, respectively) by Peltier modules (3 cm × 3 cm) driven by Wavelength Electronics PID temperature controllers. This means that the Peltier modules and correspondingly the copper plates are larger than the cell itself.

Our method of extracting the temperature and concentration from the phase shift requires only the relative value of the phase. To estimate the relative value, phase differences between the first and all subsequent measurements were calculated,  $\Delta \phi/(x, z, t_i) = \phi(x, z, t_i) - \phi(x, z, t_0)$ . The change in the refractive index  $\Delta n$  may be obtained from the unwrapped phase change  $\Delta \phi$  using the expression:

$$\Delta n = n(x, z) - n_0 = \frac{\lambda}{2\pi L} \Delta \varphi \tag{6.2}$$

Where  $\lambda$  is the wavelength and L is the thickness of the liquid in the direction of the optical pathway. For a given wavelength, the variation in the refractive index includes temperature and concentration contributions:

$$\Delta n(x, z, t) = \left(\frac{\partial n}{\partial T}\right)_{T_0, C_0} \Delta T(x, z, t) + \left(\frac{\partial n}{\partial C}\right)_{T_0, C_0} \Delta C(x, z, t)$$
(6.3)

where  $\Delta T(x, z, t)$  and  $\Delta C(x, z, t)$  are temperature and concentration variations at point (x, z).

Starting with an isothermal homogeneous binary mixture and applying a temperature gradient, a concentration profile will emerge due to the Soret effect. A steady temperature distribution will be established after a characteristic thermal time  $\tau_{th} = L^2/\chi \approx 770s$ , which depends on the size L of the experimental cell and on the thermal diffusivity  $\chi$  of the binary mixture. Simultaneously, mass separation occurs along the temperature gradient. However, mass transport is significantly slower than the thermal transport process; its characteristic time  $\tau_D$  is usually two orders of magnitude larger than the thermal time  $\tau_D = L^2/D \approx 115 \times 10^3$ , here D is the diffusion coefficient.

The stationary temperature field at the beginning of the experiment, t = 1200 s, is shown in Figure 6.2. At that time, the temperature field has already been established  $t > \tau_{th} = 770$  s, but the diffusion process had not yet begun,  $\tau_D = 115 \times 10^3$  s. After this point in time, changes in the refractive index were only due to the Soret effect-induced variations in the concentration field. At the end of the experiment, when the system was reverted back to an isothermal case, the molecular diffusion progressively reduced the previously established concentration gradient. Again, the steady temperature distribution was rapidly re-established.



**Figure 6.2:** Stationary temperature field for 90% water and 10% isopropanol mixture at the beginning of the experiment when heating from above

#### 6.5.1 Test liquids

As discussed before, in binary mixtures one can distinguish a positive Soret effect, when the lighter component is driven towards the higher temperature region (in accordance with gravity), and a negative Soret effect, when the situation is opposite (against gravity). As a result, two mixtures of water and isopropanol (IPA) of different concentrations were chosen as test fluids. The first 19 runs used the mixture of 10% IPA and 90% water, which has a negative Soret coefficient,  $S_T < 0$ . The next 20 runs used the 50% IPA and 50% water mixture, which has positive Soret coefficient,  $S_T > 0$ . Both mixtures were degassed before the cells were filled. A dedicated cell array was designed and built for each mixture, as shown in Figure 6.3. It consisted of two identical cells filled with the same liquid. The dimensions of the cell array were 8.5 cm (h) × 8 cm (w) × 30.5 cm. The primary cell contained only liquid while the companion cell also contained tracer particles for analysis of convective flows. These tracer particles were hollow ceramic microspheres with radii of 75 ± 20 µm.

The test liquids were chosen with the purpose of increasing the ratio Gs/Ra in Equation 6.6 under the condition of a minimal viscous time  $L^2/v$ , which will be described further in the next part.



Figure 6.3: Experimental setup: cell array (Primary cell, Companion cell and vibrational stimuli) [44]

#### 6.5.2 Diffusion cell

The two lateral walls are transparent and two views were available during the experiment. The advantage of two simultaneous views is beneficial for determination of the flow pattern in the convective regime. This was confirmed during parabolic flight experiments [25, 42]. A constant temperature difference ( $\Delta T = 5$ , 10 or 15 K) was maintained between the opposite horizontal walls of the cell by the Peltier elements. The mean temperature of the experiment was 25 °C. The cell was fixed to the motor that produced translational harmonic oscillations in the direction perpendicular to the temperature gradient.

#### 6.5.3 Vibrational part

The response of the fluid to external forced vibrations depends on the following factors: frequency, amplitude and the type of vibration, e.g. harmonic, rotational, or translational. The translational type of vibration was the one used in the IVIDIL experiments. Translational vibrations cause density non–uniformities, which are necessary since without them the cell with liquid inside behaves like a stone. The low and high frequencies were distinguished by comparing the oscillation period with the reference viscous and heat/mass diffusion times.

The case of small amplitude and high-frequency vibrations is of special interest. As discussed earlier, in this regime the flow field can be divided into the 'quick' part, which oscillates with the frequency of the vibration, and the 'slow,' time–averaged part (mean flow), which explains the

non-linear response of the fluid to a periodic excitation [34]. This special behaviour is shown in Figure 6.4. The amplitude of the small oscillations matches the 'quick' contribution and the displacement of the red dot represents the mean flow.



**Figure 6.4:** Particle tracing under high frequency vibration. The quick part or full motion is the blue line, and the slow time-averaged part or mean flow is the red dots [44]

The IVIDIL target was to analyze the mean flows, as they transport heat and matter in a similar way to buoyant convection. This mean flow is the reason for disturbances in the diffusive regime.

As mentioned before, the examination of the vibrational flows on the ground is masked by buoyancy. The strength of buoyant and vibrational convection is characterized by the Rayleigh number Ra and its vibrational analogue denoted by either Ra<sub>v</sub> or Gs [34]. Convection can be caused by thermal or concentration gradients. Correspondingly, one can determine thermal  $Ra_T$ or  $Gs_T$  and solutal  $Ra_C$  or  $Gs_C$  numbers defined as:

$$Ra_T = \frac{g\beta_T \Delta T L^3}{\nu \chi}, \quad Gs_T = \frac{(A\omega\beta_T \Delta T L)^2}{2\nu \chi}$$
(6.4)

$$Ra_{C} = \frac{g\beta_{C}\Delta CL^{3}}{\nu D}, \quad Gs_{C} = \frac{(A\omega\beta_{C}\Delta CL)^{2}}{2\nu D}$$
(6.5)

where g is the gravitational acceleration,  $\beta_T$  is the thermal expansion coefficient,  $\beta_C$  is the solutal expansion coefficient, v is the kinematic viscosity,  $\chi$  is the thermal diffusivity,  $\rho$  is the density, A

is the amplitude of vibration, and  $\omega = 2\pi f$  is the angular frequency.

The net relative importance of buoyant and vibration-induced convection can be roughly determined as [34]:

$$\frac{Gs}{Ra} \cong \frac{Gs_T}{Ra_T} \pm \frac{Gs_C}{Ra_C} = \frac{(A\omega)^2 (\Delta \rho / \rho)}{2gL}$$
(6.6)

where  $\Delta \rho / \rho = \beta_T \Delta T \mp \beta_C \Delta C$ . Therefore from this equation, it can be concluded that to increase vibrational effect the frequency should be high (amplitude is limited by the theory), or experiments should be performed either in a low gravity g $\approx 0$  environment or at the microscale L  $\approx 0$ . Conducting the experiments onboard the ISS satisfies one of the above-mentioned factors that can enable the comprehensive analysis of vibration-induced convection.

Also, IVIDIL experiments were performed for the frequency range of 0–2.8 Hz and the amplitudes A = 0-0.07 m. Note that the strength of vibrations depends only on the combination  $A\omega$  and the resulting mean flow through the  $Ra_v$  or Gs number. In terms of the related Rayleigh vibrations, the IVIDIL experiments were performed with values of Ra<sub>v</sub> of 0 <Ra<sub>v</sub>< 3500 for the mixture of 50% Isopropanol and 0 <Ra<sub>v</sub>< 11,400 for the mixture of 10% Isopropanol.

#### 6.5.4 Optical part

Modern techniques based on optical methods of observation do not disturb the diffusive process. Interferometry is a trusted and widely-used optical technique for measurements of the refractivity of objects. As a result, related quantities like temperature or concentration can be determined. The experimental setup was designed for observing spatial and temporal variations of composition in the mixture with the help of optical digital interferometry. This method was selected because it is considered to be the most precise method for measuring diffusion in transparent fluids. It was used for measuring thermal diffusion in about 1950 [54], but since then due to some technical difficulties, the method did achieve widespread use. An innovative part of this technique is that it allows the observation of the temperature and concentration fields across the whole cross section and not just at specific points.
In the method of Optical Digital Interferometry [41], the thermal and compositional variations are measured by the corresponding refractive index change of a diffusive media when a laser beam is passed through the whole sample. For the measurements of thermal diffusion by means of optical diagnostics, the discussed cell with transparent lateral walls that clamped between two thermostabilized blocks was used. The data were recorded by computer with a chosen sampling rate. As mentioned before, the optical portion of instrument was based on the Mach–Zehnder interferometer (Figure 6.5) in order to observe the concentration variation of the liquid mixtures. This method allowed simultaneous observation of the fringe pattern and the object itself.



Figure 6.5: The sketch of the optical method [42, 44]

A laser diode with wavelength  $\lambda = 672$  nm was employed for the IVIDIL experiment onboard the ISS. The expanded and collimated laser beam is split into the reference and objective arms with the cell assembly placed within the latter. Both beams were then redirected by mirrors and joined at the second beam splitter. The resulting interferogram was recorded by CCD camera. Resolution of imaging system was about 50 pixels/mm.

Values of refractive index variation with temperature and concentration,  $(\partial n/\partial T)_C$  and  $(\partial n/\partial C)_T$  (the contrast factors), are required for the measurement of species separation and were extracted from a set of refractometric measurements.

Although the optical measurement is the best available method in the experiment and have many advantages but there are some specific limitations for this experiment. The limitation is that some cases the accuracy of the method can change due to certain optical problems caused by the properties of the mixture. A good example to consider is water–alcohol mixtures in which there can be a large amount of scattering of data in areas of small water concentration. The reason for such data scattering is the mixing of optical signals (phase) by several factors such as concentration, optical noise, and thermal noise. Although, as will be discussed further, the basic temperature field can be taken out of consideration by subtraction of the respective reference image. In many cases, the influence of these fluctuations is negligible, but when concentration input vanishes they can become significant.

## 6.6 Digital interferometry analysis methods

Presently there are two main methods available to extract spatial distribution of optical phase from interference patterns. They are the two-dimensional Fourier transform method [55, 56] and the temporal phase-stepping technique [57]. The second one is more sensitive, but the first one has the advantage of being more robust to external disturbances. I have adopted the 2D Fourier transform method. As this method utilises the Fourier transformation of the fringe pattern, the carrier fringe system has to be sufficiently dense to provide a distinguishable peak in the Fourier domain, while at the same time, the fringe contrast should not suffer when fringes are very close together [58].

Interferograms are reconstructed by performing a 2D Fast Fourier Transform (FFT) of the fringe image, filtering a selected spectral band, performing the inverse 2D FFT of the filtered result and then phase unwrapping. Knowledge of the phase shift gives information about the gradient of the refractive index. As mentioned before, for a given wavelength, the variation of the refractive index is:

$$\Delta n(x, z, t) = \left(\frac{\partial n}{\partial T}\right)_{T_0, C_0} \Delta T(x, z, t) + \left(\frac{\partial n}{\partial C}\right)_{T_0, C_0} \Delta C(x, z, t)$$
(6.7)

where  $\Delta T(t)$  and  $\Delta C(t)$  are temperature and concentration variations at point (x, z), where the *z*-axis is the vertical direction. At the same time,  $\Delta n$  may be obtained from the phase difference  $\Delta \varphi$ , which is measured by interferometry:

$$\Delta n(x,z) = n(x,z) - n(x_0 - z_0) = \frac{\lambda}{2\pi L} \Delta \varphi(x,z)$$
(6.8)

were *L* is the optical path in the liquid. As it follows from Equation 6.8, the variation of the refractive index  $\Delta n$  is equivalent to the change of the optical phase  $\Delta \varphi$ , which is a measured value and is an additive quantity contrary to  $\Delta n$ . Herein I have used the above-described two-dimensional (2D) Fourier transform technique to calculate the phase shift  $\Delta \varphi$ . It was originally developed for treating one-dimensional (1D) phase distributions [56] and later adapted for 2D phase maps [55].

The contributions from temperature and concentration are distinguishable in time. As discussed, the beneficial part of this technique is that the measurements are performed over the entire crosssection of the cell during the whole diffusive process. It allows not only the determination of mass transport coefficients from the steady state, but also allows the observation of the evolution of the process with time and the investigation of convection when it exists.

### 6.7 Fringe analysis Steps

An example of a fringe pattern through the main processing steps is shown in Figure 6.6. After 2D Fourier transformation of the initial fringe pattern, a spectrum of fringes in the spatial frequency domain can be obtained. A typical spectrum consists of three main features (Figure 6.6): a central peak with information about the background intensity of the interferogram and two sidebands that each carries complete information about the phase. For the analysis, only one sideband from the full spectrum is required. Therefore, a Gaussian filter is used for the spectrum with the maximum located at the desired sideband.

The next and important step is then to shift the resulting spectrum towards the origin of the Fourier domain. The distance of this shift corresponds to the spatial frequency of the initial (reference) interference pattern. This operation removes the fringe carrier frequency from the spectrum and leaves only information about the change of fringe density with respect to the initial state, as well as values of the phase related to this change. In addition to the fact that the reference interferogram determines the spatial carrier frequency, it possesses its own phase distribution map, which has to be evaluated in a separate step and then subtracted from the phase map of interest. In this way, the method applies the holography principle and tracks only the posterior optical phase variation in the set of images following the reference image.



**Figure 6.6:** Principle image processing steps

By performing the inverse Fourier transform of the filtered and shifted spectrum, both the amplitude and spatial distribution of the fringe pattern phase change can be reconstructed. However, the phase difference calculated in this way is wrapped, which means that it belongs to the range  $(-\pi,\pi)$ . It should be unwrapped in order to construct the continuous natural phase. In our case, the phase maps typically were of good quality. Therefore, the simplest approach which is based on the successive comparison and validation of closed neighbour pixels was used.

This last step was performed automatically by using the proper software, a brief description follows. The procedure started at a pixel with a well-defined neighbourhood, and the assumption that an error-free phase exists. Following a spiral path, the phase unwrapping was performed by comparing the wrapped phase with that of the previously validated neighbours. If the difference

is less than  $\pi$ , the phase remains unchanged. If the difference between two pixels is more than  $\pi$ , the phase is then set equal to its wrapped phase minus  $2\pi$ . If the difference is less than  $-\pi$ , the phase is set equal to its wrapped phase plus  $2\pi$ . By the end, the relative phase change between two pixels is placed in the range  $-\pi$  and  $\pi$  and a smooth 2D phase map is obtained (Figure 6.7 a, b, c and d).



**Figure 6.7:** Phase map obtained from Fourier processing. (a) Wrapped 2D distribution and (b) wrapped vertical profile; (c) unwrapped 2D map and (d) corresponding vertical profile

## 6.8 Subtraction of reference image

The two-dimensional phase distribution (map) contains information about many aspects of the wave front. The wave front can be deformed by optics, by disturbances of air temperature, by temperature dispersal in glass walls, temperature distribution in liquid bulk, and finally by

concentration distribution in the liquid. Each particular interference pattern is formed by some or all of the above factors. An important part of the general processing is the separation of the various contributions. This segregation is done by selecting the proper reference image. For example, by choosing a reference image shortly after establishing the temperature difference over the cell, we can eliminate information about all input factors into the optical phase except for the concentration variation.

For this reason, one of the errors in the measurement came from the fact that the reference image for extracting concentration distribution was not necessarily located at the very beginning of the separation step. It had to be selected only after the temperature profile was completely established. As a result, a reliable reference image can often be found in the 10–15 minutes after the real separation has started, depending on the mixture thermal time.

Let us discuss this in more detail. The wave-front is affected and as a result distorted by optical elements along the beam path  $\Delta \varphi_{optics}$ , non-uniform air temperature  $\Delta \varphi_{air}$ , temperature distribution in glass walls  $\Delta \varphi_{glass}$ , temperature distribution in liquid bulk  $\Delta \varphi_{th}$ , and finally by the concentration distribution in the liquid  $\Delta \varphi_{c}$ :

$$\Delta \varphi = \Delta \varphi_{optics} + \Delta \varphi_{air} + \Delta \varphi_{glass} + \Delta \varphi_{th} + \Delta \varphi_{c}$$
(6.9)

The very first image was taken during the initial isothermal step. In this case, the reference image keeps information about the initial state of the optics only ( $\Delta \varphi_{ref1} = \Delta \varphi_{optics}$ ) and the subsequent phase variation was caused by possible optical path perturbations due to different mean temperatures of the air and the cell. At the end of this step, a new reference image ( $\Delta \varphi_{ref2} = \Delta \varphi_{optics} + \Delta \varphi_{air}$ ) was required to process images after applying the thermal gradient over the cell. At the second step, that had a typical duration of a few minutes, the phase variation was due to the temperature differences in the glass wall and in the liquid bulk. The concentration contribution was negligibly small at this stage as the diffusion characteristic time ( $\tau_D$ ) is much larger than the thermal time ( $\tau_{th}$ ). For the studied liquids, as mentioned before,  $\tau_D = L^2/D \approx 34$ h while  $\tau_{th} = L^2/\chi \approx 770$ s for the 90%-10% mixture, where  $\chi$  is the thermal diffusivity of the liquid. As discussed before in chapter four, these characteristic times are even higher for the 50%-50% mixture.

The optical measurement of the temperature field should be done as soon as the temperature field became established, i.e., within a few thermal times,  $t \approx 10-15$  min. Afterwards, optical measurements will start deviating due to concentration contribution. This deviation point, which appeared after the temperature difference was established, was used as the reference point for the third image processing step, i.e. thermal diffusion separation  $\Delta \varphi_{ref3} = \Delta \varphi_{ref2} + \Delta \varphi_{glass} + \Delta \varphi_{fh}$ . This new reference holds information about all input factors into the optical phase except the concentration variation. Processing the next images with respect to this reference image provided the phase change from which the full 2D map of concentration field was extracted.

Generally, this method is based on Fourier processing, and its accuracy strongly depends upon the number of periods in the analyzed signal, i.e., the number of fringes in the interferogram. Therefore, it is essential that the carrier fringe system be sufficiently dense.

#### 6.9 Development of the software

Software development refers to the two different software products used in this work: software for the image processing and numerical codes for the calculation of the governing equations describing the heat and mass transfer in the liquid under vibration.

The related in-house code was implemented as described in Chapter 3 by using the PC-SAFT equation of state. Also, as discussed in Chapter 3, the commercial software "FLUENT" was used for calculating the separation of components during the whole experiment duration, i.e. 18 hours. In this model, the Boussinesq approximation was used as described in detail earlier. In this model, the forced or controlled vibrations are applied in each run. Therefore, two different numerical models were used for calculating the concentration and temperature gradients:

- 1. Using in-house code by utilizing PC-SAFT EOS along with forced vibrations
- 2. Using the commercial Software "FLUENT" and utilizing the Boussinesq approximation along with the forced vibrations.

To follow the experimental procedure as discussed before, the numerical procedure was executed as follows:

- First, the initial temperature field over the cell was set equal to  $T=25^{\circ}C$
- Second, the temperature gradient was applied: Hot wall T=25°C +  $\Delta$  T/2; Cold wall T=25°C  $\Delta$  T/2
- The T gradient was established, i.e. Soret effect was enabled, after 5 minutes without applied vibrations
- Vibrations were then applied according to the details of each specific run for 18 hours
- Temperature was then ceased
- Diffusion was calculated during 6h

## 6.10 Experimental image processing step by step

Although the general procedure for analysis of the fringes and experimental results was discussed before, herein the details of the image processing steps used for the analysis of each particular run is described:

 By using the proper software, the image format was changed to "bmp" from the initial raw image. Also, based on the resolution of the images, it was cropped with the proper pixels (x=1920, y= 1080) to have the full images. (Figure 6.8)



Figure 6.8: Initial image, "bmp" format

2. There are two images at each specific time. The left image is the one without projector particle tracing. It can be used for concentration analysis. By using general software like Adobe Photoshop, the proper pixel numbers for image cropping of the left images were found. Four numbers were read for the left, right, top and bottom of the pictures. In order

to avoid any error, it is important to select the proper pixel numbers to exclude the walls from the cropped image. This will increase the quality of the images and therefore increase the accuracy of the related calculations.

It was important to save each set of images after each step in a new folder to avoid any overwriting of the images.



Figure 6.9: The cropped left image

3. By using the software resize.exe, all images were rotated 90 degrees clockwise.



Figure 6.10: The previously cropped image rotated +90 degree

4. By using the Fourier Phase Transform software, all images were converted to the proper form as discussed before. For this purpose, the first image was selected as a reference and one of the bright points, either in the left side or the right side, was set as a carrier. Although the initial reference can be selected from any of the early images, in order to have accurate results the final reference should be the image after one thermal time. This image was used as the reference image for the extraction of all other images in that particular run.



Figure 6.11: Fourier domain image (left) and wrapped phase image (right)

5. All the images have to be subtracted from the proper reference image. This reference, as discussed before, was selected based on the thermal time of the mixture in order to exclude the effect of temperature from the fringe interference. It was found to be better if this reference image was the same reference used in the previous step for the Fourier Phase Transform. This step was executed by writing the proper Matlab program as shown in Appendix H.



Figure 6.12: Wrapped image which was subtracted from the appropriate reference image

6. At this step all the images were rotated back to their initial position. Therefore, again by using the proper program they were rotated 90 degrees counter clockwise.



Figure 6.13: Subtracted and wrapped image rotated -90 degree to return to its original coordinate

7. As discussed before, all the images were then unwrapped in order to be able to extract the appropriate information. For this, the "PhaseUnwrape" Software was used. The output results were saved as a Matrix file and consisted of the related intensity number at each point. The "Matrix" option was selected since we wanted the value of concentration gradient between two points (near hot and cold walls) instead of the contour of the gradient.



Figure 6.14: Unwrapped phase image; the matrix file format was used for the calculations

- 8. In this step, after having all the Unwrapped matrix files, the concentration gradients of the related components (either isopropanol or water) were calculated between two points near the hot and cold walls. For this we used the proper Matlab program as shown in Appendix I, which was written in-house based on the related equations. The two points should be near the walls and the number of pixels left from the wall should be calculated based on the resolution of the mesh used in the numerical calculations. This can be done by a simple correlation between the number of pixels in the experimental images and the mesh resolution of the numerical models.
- 9. Finally all of the concentration gradient numbers were stored in a text file and the corresponding graph was drawn based on the relevant time intervals. It is important to note that the time intervals of the images are not equal. Therefore the proper time has to be extracted by writing a simple Matlab code. This code is shown in Appendix J. I found the proper time of each point by subtracting the actual date and time of the point from the date and time of the reference point. At the end, we have a set of concentration gradient numbers and their appropriate times.

There are two important notes that have to be considered during the analysis:

1. The start time of the experiment was important in order to be able to choose the proper reference image based on the thermal time of the mixture. There are some disagreements in this regard between the CSA reports, the complete set of files separated for each run and the images after the Fourier Transform step. The best method for this seems to be by examining the Fourier Transform images. One could easily find the exact image and time where the temperature was applied at the hot wall. This image has to be considered as the start of the experiment. This is the time were the temperature gradient begins to be established between the walls. Theoretically, this should take 5 minutes. Consequently, the exact time and image where the temperature was removed from the hot wall could be easily selected by looking at Fourier Transform images. This theoretically occurs after 12 hours and 5 minutes based on the experimental scenario. This will be discussed individually for each run in the next part. Generally, this disagreement can be due to

difficulties in the ISS for starting each experiment at the exact assigned time. It is important to note that all the times and dates which will be discussed further are based on GMT.

2. In general, there are two types of images for each run that were taken by two different cameras. They have to be separated and analysed separately since the pixel coordinates for cropping are different. As a result, all the steps have to be repeated for each set individually. One set of images was taken at short time intervals in order to study a short duration of time. For example, the times around 6 hours or 12 hours after the start time were particularly important for studying the mixture behaviour. The other set of images cover the whole duration of the experiment and our image processing analysis method was used to process these. These two sets of images need to separated and processed manually. They can be differentiated by looking at the images after or before the cropping step.

## 6.11 Numerical and experimental results comparison

As discussed, two different numerical models along with the experimental results were analyzed and the related graphs were drawn to compare the component separation during the 18 hour experiment time. Each run had its own frequency and amplitude, and each experiment had a start time in the ISS that was used for the image analysis as discussed before. The physical properties used in both numerical analyses obtained from the PC-SAFT EOS and Firoozabadi diffusion model.

There were nine runs in total, 8 runs in 90-10 mixture and one run in 50-50 mixture, as shown in Table 6.1. The frequencies, amplitudes and temperature differences were selected to have various Rayleigh vibration levels. Generally the 90-10 mixture was found to be more stable than the 50-50 mixture as it will be discussed further. The Figure 6.15 shows the Rayleigh vibration distribution for the assigned runs. It shows the distribution of different vibrational strengths among the selected runs. The values of vibrational strength ranged from as low as 0 or 1.27 to as high as about 4000.

Herein, I discuss the numerical and experimental results of each run. Other information required for the image analysis is also given in the related tables. The runs were sorted from the lowest vibration to the highest, as described by the Rayleigh vibration number.

Runs	Mixture water concentration	∆ <b>T (K)</b>	Frequancy (Hz)	Amplitude (mm)	Ra <sub>v</sub>
Run 2	90%	5	0	0	0.00
Run 8	90%	10	1	70	676.64
Run 9	90%	10	0.5	70	169.16
Run 10	90%	10	0.2	70	27.07
Run 11	90%	10	2	58	1858.15
Run 12	90%	10	0.05	70	1.69
Run 18	90%	15	2	57	4037.91
Run 19	90%	15	2	48	2863.45
Run 33	50%	15	0	0	0.00

 Table 6.1: Assigned IVIDIL experimental runs



Figure 6.15: Distribution of the assigned runs in terms of the Rayleigh vibration number

1. Run#2: This run was a 90-10 mixture and without any controlled vibration. Therefore, it was a good base run for studying the component separations. It was also the base run for studying thermal and mass diffusion since there was no forced vibration. Below (Table 6.2 and 6.3) are the conditions and specifications of this particular run:

Run 2					
A (mm)	f (Hz)	Ra <sub>v</sub>	$T_{hot}(K)$	$T_{cold}(K)$	$\Delta T(K)$
0	0	0	298	293	5

Table 6.2: Run#2 experimental properties and specifications

Start Temperature	Vibration Swiched on	Remove Temperature	
Gradient Establishment	(12 hour Thermal	Gradient (6 hour isothermal	End of Experiment
(about 5 min)	Diffusion)*	diffusion, Vibration on)	
Oct. 7, 2009 12:03:54	Oct. 7, 2009 12:03:54	Oct. 8, 2009 00:09:57	Oct. 8, 2009 06:09:57

**Table 6.3:** Run#2 experiment time schedule

\*In this run there is no force vibration.



Figure 6.16: Run#2 analytical, numerical and experimental results; Component separation between hot and cold walls during 18 hours

In this run since there was no vibration, the analytical solution exists and was performed to validate the results. The required code was written based on the analytical solution (Equation 6.1) in Matlab as shown in Appendix E. The results show good agreement (about 5 percent error) between the analytical solution and experimental results when diffusion coefficients obtained from experiment used in the analytical solution. Also, when diffusion coefficients obtained from PC-SAFT EOS were used in the analytical solution, the results are in good agreement (about 5 percent difference) with both the Boussinesq approximation and PC-SAFT EOS numerical results. This will be discussed further in this chapter. The agreement confirms the accuracy of

both in-house code using PC-SAFT EOS and commercial code (FLUENT) using the Boussinesq approximation.

The experimental results as shown in Figure 6.16 are higher than the numerical results of both cases, using either the Boussinesq approximation or PC-SAFT equation of state. The trends in all the cases are the same and they all show the two experimental stages, i.e. the ascending thermal diffusion and the descending molecular diffusion stages. The experimental results show more separation which can be explained by the temperature apparatus which might apply a greater temperature difference between the walls than expected, that result in more component separation. Also, as discussed in detail before, both the temperature and component separation was measured using laser images and fringe analysis method.

The analytical solution using the Soret coefficient  $S_T$  obtained from the experiment on ISS also confirms the experimental results. This clearly shows the effect of the  $S_T$  value and therefore the  $D_M$  and  $D_T$  values in the numerical analysis. In our numerical calculations, these values were obtained either from experiments performed on earth (used in FLUENT as constant values) or from the PC-SAFT equation of state and the related thermal diffusion model. The PC-SAFT EOS and Firoozabadi thermal diffusion model was also initially verified with the experiments performed on earth. Therefore, the difference in component separation between the numerical and experimental analysis can be explained. It can be concluded that the diffusion coefficients, and more importantly, the thermal diffusion coefficient of the studied mixture obtained from the ISS experiment, are larger in value than those values that were obtained from terrestrial experiments and also from the related diffusion models. However, part of the differences could be due to the experimental errors of the apparatus in the ISS as well as the related method used for laser image processing. This means that it is possible that the actual separations were not as high as those that were obtained from the laser images produced from the ISS experiments, as shown in the related figures shown in this chapter.

2. Run#12: This was the next lowest run in terms of the applied vibration. The frequency was 0.05 and the amplitude was 70 mm. The hot wall temperature was set to 303K and as a

result the  $\Delta T$  was increased to 10K. The liquid is the 90-10 water isopropanol mixture. The component separation between the two walls is shown in Figure 6.17.

Although the forced vibration that was presented in this run can reduce the component separation, due to the applied temperature differences between the walls the separation increased significantly as shown in Figure 6.17. This shows the importance of temperature and the resulting thermal diffusion in the component separation. This can be clearly seen in both the numerical and experimental results in comparison with those in Run#2 as shown in Figure 6.16. The comparison of these two graphs also reveals that component separation reaches the steady state condition faster in this run compared to Run#2 without forced vibration. This effect is clearer in the numerical graphs. This phenomenon can be due to both the effects of the higher temperature gradient between the walls and the applied vibration. The cross effect of this is that components move faster as a result of a larger driving force that ultimately reaches the maximum separation point and steady state condition more quickly.

Table 6.4: Run#12 experimental properties and specifications

Run 12					
A (mm)	f (Hz)	Ra <sub>v</sub>	$T_{hot}(K)$	$T_{cold}(K)$	$\Delta T(K)$
70	0.05	1.69	303	293	10

 Table 6.5: Run#12 experiment time schedule

Start Temperature	Vibration Swiched on	Remove Temperature	
Gradient Establishment	(12 hour Thermal	Gradient (6 hour isothermal	End of Experiment
(about 5 min)	Diffusion	diffusion, Vibration on)	
Oct. 29, 2009 09:58:09	Oct. 29, 2009 10:03:09	Oct. 29, 2009 22:07:24	Oct. 30, 2009 04:07:24



Figure 6.17: Run#12 numerical and experimental results; Component separation between hot and cold walls during 18 hours

3. Run#10: The frequency was set to 0.2 with the same amplitude as previous run, i.e. 70mm. The hot wall temperature was again set to 303K and as a result the  $\Delta T$  was also 10K. The liquid was the same 90-10 water isopropanol mixture. The numerical and experimental results for the component separation between the two walls are shown in Figure 6.18.

Comparing this run with Run#12, the effect of forced vibration can be seen. Since the temperature gradients are the same in both runs, the higher external forced vibration is the only reason that a reduction in the component separations in Run#10 versus Run#12. This effect is more significant in numerical calculations using PC-SAFT EOS. It can be concluded that in this method, the fluid mixture is affected more strongly by the applied vibration than in the other numerical method or the experimental results obtained from image processing. As a result, the component separation steadies quickly and the related  $\Delta C$  value is smaller than the other two methods.

Run 10					
A (mm)	f (Hz)	Ra <sub>v</sub>	$T_{hot}(K)$	$T_{cold}(K)$	$\Delta T(K)$
70	0.2	27.07	303	293	10

 Table 6.6: Run#10 experimental properties and specifications

 Table 6.7: Run#10 experiment time schedule

Start Temperature	Vibration Swiched on	Remove Temperature	
Gradient Establishment	(12 hour Thermal	Gradient (6 hour isothermal	End of Experiment
(about 5 min)	Diffusion	diffusion, Vibration on)	
Oct. 15, 2009 10:21:41	Oct. 15, 2009 10:26:41	Oct. 15, 2009 22:29:05	Oct. 15, 2009 04:29:05



Figure 6.18: Run#10 numerical and experimental results; Component separation between hot and cold walls during 18 hours

4. Run#9: The frequency was set to 0.5 and the amplitude the same as the previous run, i.e. 70mm. The hot wall temperature was again set to 303K and as a result the  $\Delta T$  was 10K. The liquid was the same 90-10 water isopropanol mixture. The numerical and experimental results for the component separation between the two walls are shown in Figure 6.19.

In this run, the frequency slightly was increased in comparison with the previous Run#10. There was no significant difference seen between the graphs in Figures 6.18 and 6.19.

 Run 9

 A (mm)
 f (Hz)
 Ra<sub>v</sub>
 T<sub>hot</sub> (K)
 T<sub>cold</sub> (K)
 ΔT (K)

 70
 0.5
 169.16
 303
 293
 10

Table 6.8: Run#9 experimental properties and specifications

 Table 6.9: Run#9 experiment time schedule

Start Temperature	Vibration Swiched on	Remove Temperature	
Gradient Establishment	(12 hour Thermal	Gradient (6 hour isothermal	End of Experiment
(about 5 min)	Diffusion	diffusion, Vibration on)	
Oct. 14, 2009 15:46:49	Oct. 14, 2009 15:51:49	Oct. 15, 2009 03:53:13	Oct. 15, 2009 09:53:13



Figure 6.19: Run#9 numerical and experimental results; Component separation between hot and cold walls during 18 hours

5. Run#8: The frequency was increased to 1Hz and the amplitude was the same as in the previous runs, 70mm. The hot wall temperature was again set to 303K and as a result the  $\Delta T$  was

10K. The liquid was the same 90-10 water isopropanol mixture. The numerical and experimental results for the component separation between the two walls are shown in Figure 6.20.

In this run, the applied vibration did not possess enough strength to prevent the components from separating because of the driving force caused by the temperature gradient and resulting thermal diffusion. For this reason, the component separation is almost the same as in Run#9. However, the numerical results using the Boussinesq approximation show less separation. It can be concluded that in this numerical method, the fluid mixture is more strongly affected by vibration than in the actual experiment.

Table 6.10: Run#8 experimental properties and specifications

Run 8					
A (mm)	f (Hz)	Ra <sub>v</sub>	$T_{hot}(K)$	$T_{cold}(K)$	$\Delta T(\mathbf{K})$
70	1	676.64	303	293	10

 Table 6.11: Run#8 experiment time schedule

Start Temperature	Vibration Swiched on Remove Temperature		
Gradient Establishment	(12 hour Thermal	Gradient (6 hour isothermal	End of Experiment
(about 5 min)	Diffusion	diffusion, Vibration on)	
Oct. 27, 2009 11:56:38	Oct. 27, 2009 12:01:38	Oct. 28, 2009 00:02:42	Oct. 28, 2009 06:02:42



Figure 6.20: Run#8 numerical and experimental results; Component separation between hot and cold walls during 18 hours

6. Run#11: The frequency was significantly increased to 2Hz and the amplitude of 58mm was applied for this run. The hot wall temperature was again set to 303K and as a result the  $\Delta$ T was 10K. The liquid was also 90-10 water isopropanol mixture. The numerical and experimental results for the component separation between the two walls are shown in Figure 6.21.

In this run, due to the high strength vibration applied, the separation decreased significantly in both the experimental results and numerical results. The numerical data shows a smaller magnitude than the experimental results, which confirms the previous statement that the numerical methods are more strongly affected by the vibrational effect than the actual experiment.

Run 11					
A (mm)	f (Hz)	Ra <sub>v</sub>	$T_{hot}(K)$	$T_{cold}(K)$	$\Delta T(K)$
58	2	1858.15	303	293	10

 Table 6.12: Run#11 experimental properties and specifications

Start Temperature	Vibration Swiched on	Remove Temperature	
Gradient Establishment	(12 hour Thermal	Gradient (6 hour isothermal	End of Experiment
(about 5 min)	Diffusion	diffusion, Vibration on)	
Oct. 28, 2009 15:24:25	Oct. 28, 2009 15:29:25	Oct. 29, 2009 03:30:32	Oct. 29, 2009 09:30:32

 Table 6.13: Run#11 experiment time schedule



Figure 6.21: Run#11 numerical and experimental results; Component separation between hot and cold walls during 18 hours

7. Run#19: The frequency was kept at 2Hz and amplitude of 48mm was applied. The hot wall temperature was increased to 308K to obtain a  $\Delta$ T equal to 15K. The liquid was still the 90-10 water isopropanol mixture. The numerical and experimental results for the component separation between the two walls are shown in Figure 6.22.

In this run, the vibration was increased again and as expected a decrease in the separation was observed. The experimental graph shows the mixing behaviour of the fluid mixture due to the strong vibration applied, as shown in Figure 6.22. Of interest is that the component separation changes direction twice in the first stage of the experiment at about 600 seconds.

Run 19					
A (mm)	f (Hz)	Ra <sub>v</sub>	$T_{hot}(K)$	$T_{cold}(K)$	$\Delta T(K)$
48	2	2863.45	308	293	15

 Table 6.14: Run#19 experimental properties and specifications

 Table 6.15: Run#19 experiment time schedule

Start Temperature		Vibration Swiched on	Remove Temperature	
Gradient Establishment		(12 hour Thermal	Gradient (6 hour isothermal	End of Experiment
(about 5 min)		Diffusion	diffusion, Vibration on)	
	Nov. 05, 2009 23:19:27	Nov. 05, 2009 23:24:27	Nov. 06, 2009 11:25:47	Nov. 06, 2009 17:25:47



Figure 6.22: Run#19 numerical and experimental results; Component separation between hot and cold walls during 18 hours

8. Run#18: The frequency was kept at 2Hz and the amplitude was increased to 57mm. The hot wall temperature was again kept at 308K to obtain the same  $\Delta T$  of previous run equal to 15K. The liquid was still the 90-10 water isopropanol mixture. The numerical and experimental results for the component separation between the two walls are shown in Figure 6.23.

In this run, as a result of the very strong vibration applied to the experimental cell, a strong mixing behaviour was observed, as shown in Figure 6.23. There are several directional changes in the experimental graph which is due to the strong vibration that created the strong mixing and repeatedly produces changes in the components near the walls. This behaviour can also be seen at the end of second stage of the experiment at about 960 seconds when the separation increased again and changed from descending trend to an ascending trend.

The two numerical results were closer as they show increases in the vibration. At the same time, the experimental result had a magnitude that was closer to the numerical results than in previous runs. This is due to the strong vibration that reduced the separation significantly and therefore both numerical and experimental results reach steady state quickly by reaching its maximum value.

Table 6.16: Run#18 experimental properties and specifications

Run 18					
A (mm)	f (Hz)	Ra <sub>v</sub>	$T_{hot}(K)$	$T_{cold}(K)$	$\Delta T(K)$
57	2	4037.91	308	293	15

 Table 6.17: Run#18 experiment time schedule

	Start Temperature	Vibration Swiched on	Remove Temperature	
Gradient Establishment		(12 hour Thermal	Gradient (6 hour isothermal	End of Experiment
	(about 5 min)	Diffusion	diffusion, Vibration on)	
	Nov. 05, 2009 04:43:41	Nov. 05, 2009 04:48:41	Nov. 05, 2009 16:50:33	Nov. 05, 2009 22:50:33



Figure 6.23: Run#18 numerical and experimental results; Component separation between hot and cold walls during 18 hours

9. Run#33: This was the 50-50 water-isopropanol mixture. The frequency and amplitude in this run are both zero. Therefore, in terms of applied vibration, this run was similar to Run#2 in the 90-10 mixture. The hot wall temperature was kept at 308K to obtain the  $\Delta T$  being equal to 15K. The numerical and experimental results for the component separation between the two walls are shown in Figure 6.24.

This run is similar to Run#2 but for the other mixture composition of 50-50. Generally, comparing Figure 6.24 with Figure 6.16, the component separation in the current Run#33 is greater than that of Run#2. This is due to the higher temperature gradient applied in this run in comparison with that of Run#2 with  $\Delta T$  being equal to 5K. Therefore, we can conclude that the separation is more dominant in the 90-10 mixture if it would be at an equal temperature gradient. The experimental results in this run as shown in Figure 6.24 are not very accurate and show mixing with many direction changes, as shown in the graph in Figure 6.24. This can be due to the experimental conditions at the time this particular experiment was carried out. There are many other activities in the ISS such as crews' activities, machinery and docking and undocking of the space shuttle that can alter the results. The numerical results are much closer to the

experimental results in this mixture compared to the 90-10 mixture. This can be explained by fewer differences between the diffusion coefficients calculated for this mixture in the experiments performed on earth in comparison with the ISS experiment. This refers to the Boussinesq approximation method using FLUENT. Also, the PC-SAFT EOS and Firoozabadi thermal diffusion model shows better prediction of the diffusion coefficient for this mixture in comparison with the 90-10 mixture when they are compared with the ISS experimental results.

Table 6.18: Run#33 experimental properties and specifications

Run 33					
A (mm)	f (Hz)	Ra <sub>v</sub>	$T_{hot}(K)$	$T_{cold}(K)$	$\Delta T(K)$
0	0	0	308	293	15

 Table 6.19: Run#33 experiment time schedule

Start Temperature	Vibration Swiched on	Remove Temperature	
Gradient Establishment	(12 hour Thermal	Gradient (6 hour isothermal	End of Experiment
(about 5 min)	Diffusion	diffusion, Vibration on)	
Dec. 10, 2009 18:51:36	Dec. 10, 2009 18:56:36	Dec. 11, 2009 06:57:18	Dec. 11, 2009 12:57:18



Figure 6.24: Run#33 numerical and experimental results; Component separation between hot and cold walls during 18 hours

## 6.12 Conclusion

In all the runs, the graphs of both experimental and numerical results show the same trend; however, the PC-SAFT EOS numerical calculations show more mixing and therefore less component separation. This is more significant in the runs with a higher vibrational magnitude. The numerical results utilizing the Boussinesq approximation also show more mixing and less separation in comparison with the experimental results. The reason is found to be the diffusion coefficients used in the numerical calculations which are different than that obtained from the ISS experimental results. The mass diffusion coefficient obtained from PC-SAFT EOS is slightly different than that of the ISS experimental results as shown in Table 6.20. The same applies to the thermal diffusion coefficient as shown in Table 6.20. It is also of note that in the numerical calculations using the Boussinesq approximation, the diffusion coefficients are assumed to be constant and are obtained from the PC-SAFT equation of state.

 Table 6.20: Diffusion Coefficients for Isopropanol(10%) - Water (90%) mixture

Properties	ISS Experiment	PC-SAFT EOS	
Diffusion coefficient [m <sup>2</sup> /s]	$6.31 \times 10^{-10}$	$7.77  imes 10^{-10}$	
Thermal diffusion coefficient [m <sup>2</sup> /s.K]	$4.27 \times 10^{-12}$	$2.39 \times 10^{-13}$	
Soret coefficient [1/K]	$6.77 \times 10^{-3}$	$3.076 \times 10^{-4}$	

As discussed before, the physical properties estimated by PC-SAFT EOS were already verified by Pan et al [21] in comparison with the mixture properties obtained from experiments performed in terrestrial conditions. As a result, it can be concluded that the experiments performed in the ISS zero gravity environment need more careful consideration to find the possible reasons that can affect the results.

In order to further see the effect of diffusion coefficients on the results, the diffusion coefficients obtained from PC-SAFT EOS are used in the analytical solution calculations as shown in Figure 6.26. If they are replaced by the diffusion coefficients achieved from the experimental results that were so far used in the analytical calculations, the analytical results will be quite close to the numerical results as shown in Figure 6.26. This also confirms the importance of the diffusion coefficients in the results.



Figure 6.25: Run#2 results comparison; using diffusion coefficients from PC-SAFT EOS in the analytical solution

It is also important to discuss the temperatures applied on the two lateral walls in the experiments. Although theoretically the temperatures should be exactly those noted in each run table, due to the experiment conditions, there would be some deviation. As discussed before, the temperatures are applied on the walls. The temperature of the fluid close to the walls is still not exactly what is applied on the wall plates. This was checked by using the images of the experimental results. In Run#2, the temperature difference between the two walls was monitored and is shown in Figure 6.27. It becomes steady at about 4°C which is about 1 degree less than what theoretically expected.



Figure 6.26: Temperature difference between the fluids close to the lateral walls in Run#2

If this new  $\Delta T$  (4°C) used in the analytical solution, analytical plot gets closer to the numerical calculations as shown in Figure 6.28. It is noted that based on the theory, the actual temperature difference between the fluids near the two lateral walls should be used for the calculations.



Figure 6.27: Run#2 results comparison; using diffusion coefficients obtained from experiment and actual  $\Delta T$  being equal to 4

# **Chapter 7**

## **Conclusions and Recommendations**

### 7.1 Conclusions

Molecular diffusion takes place in a mixture due to concentration gradients. Separation of the constituents in a mixture is enhanced by temperature gradients through the process of thermal diffusion. Diffusion is the result of the combined effects of temperature and concentration. The density gradient also may result from non-uniformity in temperature or composition. The thermal diffusion (the Soret effect) phenomenon is a coupling between a temperature gradient and its resulting mass flux in a multi-component system. A steady state is reached when the separating effect of thermal diffusion is balanced by the remixing effect of molecular diffusion which occurs due to a concentration gradient in a mixture that tends to decrease the concentration gradient. Thermal diffusion has a significant effect in many important processes in nature and technology. Thermohaline convection in oceans [1], in the analysis of the distribution of components in oil reservoirs [2], isotope separation in liquid and gaseous mixtures [3] and characterization and separation of polymers [4].

Vibrational convection refers to the specific flows that appear when a fluid with a density gradient is subjected to external vibration. Experiments onboard the International Space Station experience a convective flow due to the oscillatory vibrations called g-jitters induced from various sources such as crew activities, mechanical systems, thrusters firing, spacecraft docking, etc. Although g-jitter seems to have a major impact on diffusion-related experiments in Space, very few experimental studies have addressed this topic. In these experiments, the results did not provide clear evidence of time-averaged flows nor of the related heat transfer caused by periodic high-frequency g-jitter. Therefore, it was an essential step to further investigate the effect of these oscillations in the fluid mixtures. The outcome can be very useful for the improvement in the future experiments on space to further improve the accuracy. It can also be used in possible applications of thermovibrational convection in the fluid transport at the zero gravity environment of space. In this study, the transport processes (fluid flow, heat transfer and mass

transfer) in the presence of the oscillatory g-jitters were studied in a set of thermal diffusion experiments carried out in the International Space Station.

In this work, extensive numerical modelling was performed and the experimental results obtained from the ISS were also analyzed in detail. The results were compared based on the theoretical, numerical and experimental work carried out, and the following conclusions can be made:

 The water and isopropanol mixture with two different compositions was selected in order to have both positive and negative Soret coefficients. Three scientific groups were involved in the benchmark study. In order to simplify the problem, the fluid was modelled as a single fluid instead of a binary mixture. Therefore, the physical properties of the mixture were used and consequently the effect of diffusion coefficients was not included in the analysis at this stage.

The benchmark results show that establishment in both temperature and velocity is faster in the cases with non-adiabatic vertical walls and the thermovibrational convection is slightly stronger in those same cases. In summary, three main conclusions were made from the graphs:

- a. Generally thermovibrational convection is stronger in the 50%-50% mixture in comparison with 90%-10% mixture both in the adiabatic and non-adiabatic case.
- b. The non-adiabatic cases, i.e. linear temperature distributions along the vertical wall in both of the mixtures (50%-50% & 90%-10%), have stronger thermovibrational convection as of the higher Nusselt numbers obtained.
- c. The 90%-10% mixture has faster establishment of steady state for both temperature and velocity in comparison with the 50%-50% mixture in both the adiabatic and non-adiabatic cases. In another words, this behaviour can also be supported by the stronger thermovibrational convection experienced in the 50%-50% mixture in comparison with the 90%-10% mixture that delays the establishment of a steady temperature and velocity in this mixture.

In addition, similar results were obtained using both micro gravity  $(10^{-6}g)$  and zero gravity for the analysis. In spite of good agreements achieved between the groups, it was decided to use the mean flow approach for the second benchmark analysis. This was due to the difficulties in analyzing the instantaneous snapshots of the flow because of the quickly changing flow due to the effect of applied vibrations.

2. This benchmark study was performed on the identification of the vibrational convection intensity and enhancement of heat transfer. The output of this study was used for preparation and explanation of the experimental results described in Chapter 6. In this study, eight different levels of frequency and amplitude were applied to the model that was already filled with the two mixtures.

The behaviour of the flow due to vibration was studied and after some iteration, complete agreement was achieved between all the scientific groups. It was found that the mean flow pattern is the most reliable way to compare the flow behaviour. As suggested by the Gershuni theory [34], in all runs four vortex patterns were obtained since the Rayleigh vibrations are all below the critical value [34]. The evolution of the flow at different characteristic times was studied. It was found that in the low vibration intensity regime, i.e. low Rayleigh vibration, the four vortex pattern can be created at a later time in comparison with the higher Rayleigh vibration regime. The Nusselt numbers near the walls and the average velocity over the entire domain was studied for all vibrational intensities in both the adiabatic and non-adiabatic cases at various characteristic times. The trend was an increase in the Nusselt number from runs with low Rayleigh vibration to those with high Rayleigh vibration. Almost no significant difference was found between adiabatic and non-adiabatic cases. However, in some runs, average velocities and Nusselt numbers were higher in non-adiabatic cases in comparison with the adiabatic. This is in agreement with our conclusion for the first benchmark study that thermovibrational convection is stronger in the non-adiabatic cases.

3. Double diffusive convection in a binary mixture of isopropanol (10%) and water (90%) at zero gravity condition in the presence of Soret effect was studied. This effect created a

concentration gradient due to the temperature gradients between the walls. The direction of the displacement for each component depends on the sign of the Soret coefficient and for the current mixture the Soret coefficient is negative, which results in high concentration of water in the hot side of the cavity.

The external vibrations applied in the model to create thermovibrational convection. As a result, an increase in the heat transfer and a decrease in the concentration gradient were produced. This is observable in the zero gravity condition due to the absence of buoyant convection. The component separation or Soret separation and the maximum temperature deviation from the ideal conditions were also studied. These two parameters were used to indicate the significance of the effect of convection induced by vibration on diffusion. The structure of the time-averaged (mean) fields was examined on a long time scale, i.e., during one thermal time along with the composition and temperature profiles. Three different levels of Rayleigh vibrations were considered by using different frequencies, amplitudes, and temperature gradients. Two models, the weighted average mixing rule and PC-SAFT EOS were used for calculating the density.

The one diagonal mean flow pattern was seen in Rayleigh vibrations less than the critical number, as Gershuni theory predicted. Therefore, the velocity magnitude is higher when the density is calculated using PC-SAFT EOS. Accordingly, significant mixing occurs when using PC-SAFT EOS that results in less component separation. It was found that this is due to coupling between the temperature and density in PC-SAFT EOS that makes higher instantaneous changes in the density. This will result in higher velocity magnitudes which then are reflected in component separations.

4. The experiment IVIDIL (Influence of Vibrations on Diffusion in Liquids) was performed in the Fall of 2009 onboard the ISS, inside the SODI (Selectable Optical Diagnostics Instrument) mounted in the glove box on the ESA Columbus module. It carried out 41 experimental runs with each of them lasting 18 hours. Three international teams were involved in the project. Other than our team in Ryerson University led by Dr. Saghir, a Team from Université libre de Bruxelles was responsible for aspects related to IVIDIL experimental definition, theoretical and numerical modelling and a Russian team from Perm University provide theoretical and numerical support. Nine runs were dedicated for our group to analyze the experimental results using image processing tools and for comparison with the related analysis as well as the numerical analysis using the two approaches described in previous chapters. The objectives of the experiment, numerical analysis and the related comparisons were:

- a. After each space experiment, there was a discussion about the role of onboard g-jitters. The experiment helped to identify the limit level of vibrations where g-jitter does not play a role for onboard experiments. This was satisfied by observing the diffusive process under different imposed and controlled vibrations.
- b. To perform accurate measurements of diffusion and thermal diffusion coefficients for two binary mixtures in the absence of buoyant convection. The measured values can be used as standards for ground experiments. Two different concentrations of water– isopropanol (IPA) with positive and negative Soret effect were used as test fluids. This objective is also related to the influence of vibrations on the measured values of diffusion and thermal diffusion coefficients.
- c. Finally, to investigate vibration-induced convection and, particularly, heat and mass transfer under vibrations.

In all the runs, the same trend was achieved for analytical, numerical and experimental results, however the PC-SAFT EOS numerical calculations show more mixing and therefore less component separation. This was more significant when the vibrational intensity was higher, i.e. a bigger Rayleigh vibration number. The numerical results utilizing the Boussinesq approximation also show more mixing and less separation in comparison with the experimental results. The reason was found to be the diffusion coefficients used in the numerical calculations which were different from the experimental results. The mass diffusion coefficient obtained from PC-SAFT EOS was slightly different than that from the experimental results. The difference in the thermal

diffusion coefficient was more significant to the results. To confirm the effect of the diffusion coefficients, the analytical calculations and numerical calculations were repeated by using diffusion coefficients obtained from the experiment. The importance of using accurate diffusion coefficients especially thermal diffusion coefficient was verified.

Also, the effect of temperature gradient applied between the walls in the experiment was studied. It was found that the accuracy of the temperature gradient is important in the analysis of components separation using numerical calculations. Therefore, applying an accurate temperature gradient that was used in the experiment to the numerical model has an important role on the accuracy of the results.

## 7.2 Contributions

The main goal of this work was to investigate numerically and experimentally the effect of vibration on the diffusion in an ideal zero gravity environment of International Space Station where the effect of buoyancy convection is minimized. The appropriate mixtures of water and isopropanol at two different concentrations were selected and subjected to various levels of vibrations numerically using both the Commercial Software FLUENT and an in-house code. Extensive work was carried out in the numerical analysis to harmonize the model and calculations between all the groups involved prior to the experiment. The exact same scenario was modelled and calculations performed in the transient base. The experiments were performed at ISS and the results were analyzed and compared with the numerical analysis. Below are my contributions in this project that were published and presented in the related journals and conferences [60-64]:

- The numerical model was made using the Commercial Software FLUENT and the best approach to perform the numerical calculations in the vibrational effect on diffusion process was investigated in a set of analyses carried out in contribution with other scientific groups in Europe.
- The selected approach was tested in a series of benchmark numerical analyses performed with other scientific groups to investigate the identification of the vibrational convection
intensity and enhancement of heat transfer. The output of this study was used for preparation and explanation of the experiment performed later at ISS. It was found that the mean flow pattern is the most reliable way to study and compare the flow behaviour.

- Two different numerical models were compared for the studied mixture and subjected to different levels of vibration. The fluid flow behaviour along with the concentration gradient in the presence of vibrational intensity and the Soret effect is studied in detail. A distinctive feature was found in the behaviour of the flow due to the thermovibrational convection in comparison with the related established theory in this subject.
- The experimental results, in the form of laser images obtained from an optical digital interferometry experiment performed on the ISS, were analyzed. The results were compared with two different numerical approaches, the Boussinesq approximation and a mixing rule using commercial software FLUENT and PC-SAFT EOS and Firoozabadi Model using an in-house code. The related in-house code was developed and tuned in comparison with the available ground base experimental results. It is an experiment independent model in terms of the input values i.e. physical properties and diffusion coefficients. A set of unique graphs were produced from the analysis of the experiments on the ISS and the related numerical calculations that modeled the same scenario. The results are very useful for future scientific ISS experiments in terms of reliability of the experimental results from space and to select the best numerical approach for the comparisons.

#### 7.3 Future work and recommendations

There are still some related open issues that need to be studied, so future work may be carried out to address the followings:

 Numerical analysis of the effect of vibration on the diffusion process using another Equation of State instead of PC-SAFT. There are other Equation of States available in the literature that could better predict the physical properties. It would be very useful if they replaced in the numerical calculations and the results compared with the results obtained from ISS experiments.

- Numerical analysis using other diffusion models instead of the studied Firoozabadi model. In parallel to replacing the Equation of State in the numerical calculations, it is also essential to adopt other diffusion models. This is important as it was found that the most important factor in the results obtained from numerical calculations is the effect of Soret coefficient and most importantly thermal diffusion coefficient. It is possible that PC-SAFT EOS prediction of the physical properties was enough accurate but the diffusion model cannot match the space experiment results in terms of diffusion coefficients.
- Numerical analysis using different models for calculating the physical properties such as density, viscosity and etc. There are also many models for calculating physical properties in the literature. They can be replaced by the mixing rules used for density and viscosity calculations. This applies in the numerical calculation using FLUENT software.
- Investigate the experimental procedure and devices used in the IVIDIL experiment at ISS to study the accuracy of the results. As discussed, it is important to investigate the possible errors in the ISS experiments. This includes the apparatus used in the experiments such as for applying temperature and also external effects that can have negative influence in the space related experiments.
- As discussed before, there are some differences between the experimental results obtained from ISS experiments and the similar previous ground base experiments. It is important to perform a comparison between the results in order to find the possible improvements in the future similar experiments. It is also very beneficial to perform numerical analysis using the results obtained from both experiments i.e. previous ground base and new ISS experiments as constant values and comparing the final numerical results. This will be very useful to adopt the best numerical procedure for the future analysis.
- Revise the analysis approach of the experimental images with a better optimization model to improve the accuracy of the results.
- Perform numerical calculation using 3D model to have more realistic results in comparison with the actual experiment.

#### Appendix A

### User Defined Function for temperature distribution in nonadiabatic walls

```
#include "udf.h"
DEFINE_PROFILE(temperature_profile, thread, position)
{
  real r[ND_ND];
  real y;
  face_t f;

  begin_f_loop(f, thread)
  {
    F_CENTROID(r,f,thread);
    y = r[1];
    F_PROFILE(f, thread, position) = 293+1000*y;
  }
  end_f_loop(f, thread)
}
```

#### **Appendix B**

# User Defined Function for Boussinesq approximation with thermal expansion coefficient

In this UDF only thermal expansion coefficient ( $\beta_T$ ) take into account and the applied vibration have the sinusoidal form with the desired frequency and amplitude:

```
#include "udf.h"
#define TGr 293
#define BETAt m 0.00031
#define w 3.1415
#define a 0.07
DEFINE_SOURCE(G_L,c,t,dS,eqn)
{
 real x[ND ND];
 real Source;
 real Boussinesq;
 real Go;
  real time;
 Boussinesq = C R(c,t) * (BETAt m*(C T(c,t)-TGr));
 time = CURRENT TIME;
 Go = -a^*w^*w^*\cos(w^*time);
  Source = (Boussinesq * Go);
 dS[eqn] = 0;
 return Source;
}
```

#### Appendix C

### User Defined Function for Boussinesq approximation with thermal expansion coefficient at microgravity environment

In this UDF thermal expansion coefficient ( $\beta_T$ ) is counted only and there is no sinusoidal vibration involved. But the gravity assumed to be micro gravity i.e.  $g = 10^{-6} m/s^2$ 

```
#include "udf.h"
#define TGr 293
#define BETAt_m 0.00031
#define Go 0.000001

DEFINE_SOURCE(G_L1,c,t,dS,eqn)
{
   real x[ND_ND];
   real Source;
   real Boussinesq;

   Boussinesq = C_R(c,t)*(BETAt_m*(C_T(c,t)-TGr));
   Source = (Boussinesq * Go);
   dS[eqn] = 0;
   return Source;
}
```

#### **Appendix D**

### User Defined Function for Boussinesq Approximation with both thermal and solutal expansion coefficient

In this UDF both thermal expansion coefficient ( $\beta_T$ ) and solutal expansion coefficient are counted and the applied forced vibration had the sinusoidal form with the desired frequency and amplitude:

```
#include "udf.h"
#define TGr 293
#define BETAt m 0.00031
#define w 0.31415
#define a 0.068
DEFINE SOURCE(G L,c,t,dS,eqn)
 real x[ND ND];
 real BETAc[1]={0.1386};
 real rho[1]={998.2};
 real Mw[1]={18.0152};
 real Source;
 real Boussinesq;
 real Go;
 real time;
  int i;
 real Con = 0;
  for (i=0; i<1; i++)
    {
      Con = Con + BETAc[i]*((C_YI(c,t,i)*rho[i]/Mw[i])-(0.9*rho[i]/Mw[i]));
    }
  Con = Con + (BETAt m^*(C T(c,t)-TGr));
  Boussinesq = (C R(c,t) * (1-Con));
  time = CURRENT TIME;
  Go = -a*w*w*cos(w*time);
  Source = (Boussinesg * Go);
 dS[eqn] = 0;
 return Source;
}
```

#### **Appendix E**

#### MATLAB code for analytical solution

```
function Analytical
clear all;
close all;
clear
clc
CO=0.5; % initial concontration in mass fraction
%Dt=3.98e-12;
                % Molecular diffusion coefficient
Dm=6.27e-10;
S1=9.24e-3;
                % Soret coefficient
S=C0*(1-C0)*S1;
           % height of the cell along vertical direction
L=0.005;
DelT=4;
          % temperature difference
td=6*60*60; % time to reach steady state condition
tend=12*60*60; % total time
z1=1:1:1024;
               % number of pixels in vertical direction
               % length of every pixels in meter
z=z1/1024*L;
%%% Step-1
for i =1:length(z)
    for t=0:60:td
        sum =0;
        for n=1:2:101
            sum=sum+1/n^2*cos(n*pi*z(i)/L)*exp(-n^2*pi^2*Dm*t/L^2);
        end
        C(i,t/60+1)=C0+S*DelT*(0.5-z(i)/L-4/pi^2*sum);
    end
end
DelCst=C(i-5, end)-C(5, end);
%%% step-2
for i =1:length(z)
    for t=td+60:60:tend
        t2=t-td;
        sum =0;
        for n=1:2:101
            sum=sum+1/n^2*cos(n*pi*z(i)/L)*exp(-n^2*pi^2*Dm*t2/L^2);
        end
        C(i,t/60+1)=C0-DelCst*4/(pi^2)*sum;
    end
end
for t=0:60:tend
    t1=t/60+1;
    DelC(t1) = C(i-5, t1) - C(5, t1);
    DelC(t1) = abs(DelC(t1));
    fid=fopen('DelC-analytical.txt','A');
           fprintf(fid,'%10E',DelC(t1));
                fprintf(fid, '\r\n');
                     fclose(fid)
end
```

```
t=0:60:tend ;
plot(t/60,DelC,'o')
title('Analytical Concentration difference Vs time plot')
xlabel('Time (minute)')
ylabel('Delta C')
```

#### Appendix F

#### MATLAB code for mean velocity and temperature contours

```
clear all;
close all;
A=zeros(5202,1); % null vector
for k = 1.53845E+03:0.05:1.5409E+03
  data. filename = sprintf('data. %6.5E', k);
  asddata = load(data. filename);
A=A+asddata;
end
A=A/50;
for i=290:578
    Vx=A(i);
end
fid=fopen('Vx.txt','Vx');
       fprintf(fid,'%10E\n',A);
                  fclose(fid)
for i=579:867
    Vy=A(i);
end
fid=fopen('Vy.txt','Vy');
      fprintf(fid,'%10E\n',A);
                  fclose(fid)
for i=1157:1445
    T=A(i);
end
fid=fopen('T.txt','T');
       fprintf(fid,'%10E\n',A);
                  fclose(fid)
```

#### Appendix G

### MATLAB code for preparing required file for mean velocity vector profile animation in TECPLOT

```
clear all;
close all;
A=zeros(5202,1); % null vector
for s = 5:5:800
        A=0;
for k = s:0.05:s+2.45
  data. filename = sprintf('data. %6.5E', k);
  asddata = load(data. filename);
A=A+asddata;
end
A=A/50;
  B=load('mesh17.txt');
  for q=1:289
      X(g) = B(g, 1);
  end
  for g=1:289
      Y(g) = B(g, 2);
  end
for i=290:578
    Vx(i-289)=A(i);
end
    for m=579:867
        Vy(m-578) = A(m);
    end
        for n=1157:1445
            T(n-1156) = A(n);
        end
    fid=fopen('RUN18-INC5-TEC-MOVIE.txt','a');
        fprintf(fid, ' \n');
        fprintf(fid, 'title = "velocity vector, temperature contour"\n');
        fprintf(fid,'variables = "x", "y", "Vx", "Vy", "T"\n');
        fprintf(fid,'zone i=17, j=17,\n');
        fprintf(fid, ' \n');
for i=1:289
        fid=fopen('RUN18-PC-INC5-TEC-MOVIE.txt','a');
           fprintf(fid,'%10E\t\t %10E\t\t %10E\t\t %10E\t\t
%10E\n',X(i),Y(i),Vx(i),Vy(i),T(i));
                  fclose(fid)
end
end
```

#### **Appendix H**

## MATLAB code for image processing and subtracting the desired two images

```
function subtraction_thermodiffusion
Fref = ['Int0001PHA.bmp'];
Iref = imread(Fref, Fref(end-2:end));
Iref = im2double(Iref);
Iref = Iref*2*pi;
for k = 1000:1:2504
    I = sprintf('Int%gPHA.bmp', k);
% I = ['Int0250PHA.bmp'];
I=imread(I,I(end-2:end));
I = im2double(I);
I=I*2*pi;
nIcf=sprintf('Int%gPHA-Int0001PHA.bmp',k)
I=I-Iref;
I(I < 0) = I(I < 0) + 2*pi;
%figure
%imshow(I,[0 2*pi]),
imwrite(mat2gray(I),nIcf,'bmp')
end
end
```

#### **Appendix I**

# MATLAB code for image processing and calculating $\Delta C$ between top and bottom walls

```
function calculation_dc_top_bottom
```

```
lambda=635*1e-9;
depth=0.01;
dndC= 0.092305;
centralpixel=754/2;
w1m=0.1; %initial concentration
%unplotted distance from bottom/cold wall
z1=(15*0.01/785)+(0.5*0.01/785);
%unplotted distance from top/hot wall
z2=0.01-(15*0.01/785)-(0.5*0.01/785);
L=linspace(z1, z2, 754);
%sL=size(L)
%L=Linter(1:318);
%Linter=linspace(7.8864e-6,0.005-7.8864e-6,317);
%L=Linter(1:304);
for cpt=1000:1:2505
    name1=sprintf('Int%4dPHA-Int0001PHA.txt',cpt);
    UPhi1=dlmread(name1);
    %UPhicropped=UPhi1(:,3:end-4);
    %for i=1:1:x
        %UPhi1moyen=mean(UPhicropped);
        UPhi1moyen=mean(UPhi1,2);
    %end
    n(cpt,:)=UPhi1moyen*lambda/(2*pi*depth);
    %sn=size(n)
    wle(cpt,:)=n(cpt,:)/dndC;
    wle(cpt,:)=wle(cpt,:)-wle(cpt,centralpixel)+wlm;
    %sw=size(w1e)
    dw(cpt) = wle(cpt, end-5) - wle(cpt, 5);
    dw = abs(dw(cpt))
    fid=fopen('DelC.txt','A');
       fprintf(fid,'%6E',dw);
            fprintf(fid, '\r\n');
                fclose(fid)
```

end

#### **Appendix J**

#### MATLAB code for extracting the time of images

```
clear all;
close all;
files = dir('*.bmp');
for i = 1:numel(files)
numberStr = regexp(files(i).name,...
'img_(\d*)_(\d*)_bmp','tokens');
numberStr = numberStr{1};
numberStr = numberStr{1};
s = number
fid=fopen('numbers.txt','A');
fprintf(fid,'%g %g %g \r\n',S);
fclose(fid)
```

end

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