Thermoacoustic Convection and Transport in Supercritical Fluids under Normal and Micro-gravity Conditions

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ABSTRACT

Thermoacoustic Convection and Transport in Supercritical Fluids under Normal and Micro-gravity Conditions

Zhiheng Lei

Bakhtier Farouk, Ph. D

The generation, propagation and dissipation of thermally induced and mechanically driven acoustic waves in supercritical nitrogen and carbon dioxide were studied. Supercritical fluids are widely used in various industrial and laboratory processes as substitutes for organic solvents. These fluids also have high compressibilities, high thermal conductivities, low viscosities, and low thermal diffusivities. As the thermal diffusivity tends to zero near the critical point, acoustic waves are believed to be the primary reason for fast thermal equilibration in supercritical fluids. The compressible form of Navier-Stokes equations for Newtonian fluids was considered to model the supercritical fluids. A high-order explicit numerical scheme (FCT: flux-corrected transport, along with an accurate wall density boundary condition, was applied to accurately track the acoustic waves. The property variations (pressure, internal energy, viscosity, and thermal conductivity) of the supercritical fluids (carbon dioxide and nitrogen) as functions of temperature and density were obtained from the NIST Standard Reference Database 12 instead of simplified equations for real gases. Both one-and two-dimensional computational fluid dynamics models were developed for predicting the temporal evolution of pressure, density, temperature and flow field in supercritical fluids due to thermal and mechanical excitations. The flow fields and heat
transport induced by thermally induced acoustic waves and buoyancy in supercritical fluids were investigated numerically under normal and reduced gravity conditions. The flow fields induced by mechanically driven acoustic waves in an enclosure driven by a vibrating wall were also numerically studied. The model developed was also used to investigate the interaction of thermally induced acoustic waves and mass transfer in supercritical fluids (naphthalene dissolution in supercritical carbon dioxide).

The generation and propagation of thermally induced acoustic waves due to rapid heating of a solid wall in a confined supercritical fluid layer were also experimentally investigated. A high-pressure experimental facility was constructed to characterize the generation and decay of acoustic waves in supercritical nitrogen and supercritical carbon dioxide in an enclosure due to rapid heating of an end wall. The underlying physics of the ‘piston effect’ (fast thermal equilibration of supercritical fluids with high compressibilities) were studied and explained in detail.

To the author’s knowledge, this is the first attempt where detailed simulations of thermally induced and mechanically driven acoustic waves in supercritical fluids have been conducted with accurate equations of state and property functions. Better understanding of these problems will help the tailoring and optimizing the operating conditions for industrial and laboratory processes for supercritical fluids.
CHAPTER 1
INTRODUCTION

1.1 Overview

The investigation described in this thesis concerns the interaction between heat and acoustic energy, and the effect of the induced acoustic waves on the transport in highly compressible fluids (supercritical carbon dioxide and supercritical nitrogen) with convection / diffusion. The thermo-mechanical conversion of acoustic wave energy to heat, which is significant near the critical points, exhibits a pronounced effect on the induced flows in supercritical fluids with and without gravity. Due to the special properties of supercritical fluids, they are widely used in industry. Thus studying the heat transfer and transport in supercritical fluids has both intrinsic interest and underlying importance. The focus of this research is in carrying out numerical simulations and experimental observations of thermally induced and mechanically driven acoustic waves in supercritical fluids and in explaining their effects on heat transfer and transport.

1.2 Thermally Induced and Mechanically Driven Acoustic Waves in Fluids

Acoustic waves are generated by various kinds of sources. Usually acoustic waves are generated by mechanical oscillations, for example the vibrations of solid surfaces. They can also be induced by some thermal effects, such as rapid heating or cooling, combustion, special arrangement of heat source and sink, and so on.
When a compressible fluid is subject to a rapid temperature increase along a solid wall, the fluid in the immediate vicinity of the wall is heated by conduction, and tends to expand. The inertia of the unperturbed fluid constrains the sudden expansion due to the sudden energy input, which induces a pressure wave called thermally induced acoustic wave [1,2]. The thermally induced acoustic wave travels at approximately the local speed of sound within the fluid, and is reflected from the other wall when impinging on it. The wave repeatedly traverses the length between both walls, and its amplitude eventually damps out due to the viscous and thermal losses within the fluid.

The weak fluid velocities induced by the acoustic wave may significantly enhance heat and mass transfer within the fluid. Consequently, thermal equilibrium in the fluid is reached more rapidly than it would have been by pure conduction. This effect may be very significant near the critical point [3], or when other modes of convection are weak or absent, such as under zero-gravity conditions. Due to the high density and compressibility, acoustic waves are much stronger in supercritical fluids than in ideal gases. Furthermore the thermal diffusivity tends to be zero at the critical point; the effect of these acoustic waves on heat transfer in supercritical fluids becomes significant near the critical point, when the system undergoes a small temperature variation.

For intense sound waves that are driven mechanically, one of the challenging problems is the interaction of acoustic waves and solid boundaries. It is well-known that sound sources may generate an acoustic field in which the particle velocities are not simply sinusoidal, and a pattern of time-independent vortical flows or steady circulations is often found in the body
of compressible media [4]. These second-order steady flow patterns are known as acoustic streaming, which always have rotational character. The streaming velocity increases with the sound intensity.

Acoustic streaming may be effective in accelerating certain kinds of rate processes and has applications in localized micro-mixing, convective cooling, acoustic compressors, micro-fluidic devices, acoustic levitation, ultrasonic medical diagnostic devices and ultrasonic cleaning of contaminated surfaces.

1.3 Introduction to Supercritical Fluids

A fluid is supercritical, when its temperature and pressure are above its critical values. Figure 1.2 shows carbon dioxide phase diagram. In Figure 1.1, carbon dioxide is supercritical in the blue region, where its pressure and temperature are above its critical pressure (7.3773 MPa) and temperature (304.13 K).

Figure 1.1 Carbon dioxide phase diagram
A supercritical fluid is neither a gas nor a liquid, and exhibits a number of specific properties, such as large density, high thermal conductivity, high specific heat, low viscosity, low thermal diffusivity and low sound speed, which make it an intermediate state between gases and liquids. Comparisons of typical values for density, viscosity and diffusivity of gases, liquids, and supercritical fluids are presented in Table 1.1. Additionally, close to the critical point, small changes in pressure or temperature result in large changes in density, allowing many properties to be "tuned".

There is no surface tension in a supercritical fluid, as there is no liquid/gas phase boundary. By changing the pressure and temperature of the fluid, the properties can be “tuned” to be more liquid or more gas like.

Table 1.1 Comparisons between typical gas, supercritical fluid and fluid properties [5]

<table>
<thead>
<tr>
<th>Property</th>
<th>Density (kg/m$^3$)</th>
<th>Viscosity (µPa·s)</th>
<th>Diffusivity (mm$^2$/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gases</td>
<td>1</td>
<td>10</td>
<td>1-10</td>
</tr>
<tr>
<td>Supercritical Fluids</td>
<td>100-1000</td>
<td>50-1000</td>
<td>0.01-0.1</td>
</tr>
<tr>
<td>Liquids</td>
<td>1000</td>
<td>500-1000</td>
<td>0.001</td>
</tr>
</tbody>
</table>

1.4 Motivation for Present Research

Reviewing past literature indicates that most of the numerical work for thermally induced acoustic waves under zero-gravity condition is done in ideal gases. Due to the
specific thermophysical properties of supercritical fluids, thermally induced acoustic waves are stronger in supercritical fluids than in ideal gases, and play an important role in heat transfer problems in supercritical fluids. The extension of study of thermally induced acoustic waves to the supercritical fluids is considered in this work.

Heat transfer in supercritical fluids exhibits an interesting phenomenon: the piston effect [6-8], due to the specific thermophysical properties of supercritical fluids. Heat transfer problems in supercritical region now are attracting increased research attention. However most of the published research work is based on some simplified (and sometimes erroneous) assumptions about the thermophysical properties. For example in several studies internal energy of supercritical fluid is considered as a function of temperature only whereas it is a function of both temperature and pressure (or density). Thus it is worthwhile to study heat transfer and thermally induced acoustic waves in supercritical fluids together, with accurately representing thermophysical properties.

The experimental work on thermally induced acoustic waves is limited, even for ideal gases. So far there is no direct experimental measurement of thermally induced acoustic pressure waves in high-pressure fluids and in supercritical fluids. There are, however, indirect experimental work about thermally induced acoustic pressure waves in this region, using interferometry [9].

Natural convection heat and mass transfer in supercritical fluids have rarely been studied with the effect of thermally induced acoustic waves in the past [10,11]. It is noted
here that supercritical fluids are widely used as good solvents (for mass transfer) in industry [12-14], the most familiar example is the decaffeination of coffee beans and green tea by supercritical carbon dioxide [15,16]. For long range space flights supercritical fluids have been considered for reclaiming potable water from the urine of the astronauts[17]. Detailed numerical simulations are needed to clearly understand the physics of natural convection, heat and mass transfer in supercritical fluids with and without the effects of thermally induced acoustic waves.

In the area of mechanically driven acoustic waves, most studies are limited to ideal gases [18-20]. As supercritical fluids are widely used in industries and attract more and more research interest, it is worthwhile to extend the study of mechanically driven acoustic waves to supercritical regions.

1.5 Research Objectives

In this study, acoustically driven flow fields, heat and mass transfer in supercritical fluids, are investigated via numerical and experimental methods. Both thermally induced and mechanically driven acoustic waves are considered. Following problems are studied:

- Numerically simulate the generation, propagation and dissipation of thermally induced acoustic waves in supercritical fluids. The physical mechanisms of piston effect and heat transfer in supercritical fluids are investigated.
• Numerically study the interaction of thermally induced acoustic waves and natural convection in supercritical fluids. The acoustic waves, induced flow fields and Nusselt numbers are investigated under different initial conditions and heating rates.

• Generate and measure thermally induced acoustic waves in a high pressure and heated experimental test cylinder filled with either nitrogen or carbon dioxide. The fast variations of wall temperature are also measured to study the effect of heating rate on thermally induced acoustic waves.

• Numerically investigate the acoustic streaming, resulting from mechanically driven acoustic waves, in supercritical fluids.

• Numerically study the heat and mass transfer in supercritical fluids due to the thermally induced acoustic waves. The dissolution of naphthalene in supercritical carbon dioxide is considered.

1.6 Outline of the Thesis

The remaining chapters the thesis are organized as follows:

In Chapter 2, literature is reviewed for past experimental and numerical studies of thermally induced acoustic waves in ideal gas, liquids and supercritical fluids. The thermophysical properties and applications of supercritical fluids are then discussed. Past
heat and mass transfer studies in supercritical fluids are then discussed. In Chapter 3, the numerical model developed for describing acoustic waves and boundary conditions is presented. The numerical scheme employed including the treatment of boundary conditions is then discussed.

Chapter 4 presents the numerical results of one-dimensional thermally induced acoustic waves in supercritical fluids. The physical mechanisms of piston effect, which is a particular heat transfer mode in supercritical fluids, are then discussed. Numerical study of two-dimensional natural convection in supercritical fluids is investigated in Chapter 5. The short-time and long-time numerical scheme are also discussed. Chapter 6 presents the experimental apparatus and procedures, along with the presentation of experimental results.

Chapter 7 focuses on the flow fields created by mechanically driven acoustic waves in an enclosure filled with supercritical carbon dioxide, along with its effect on energy transport. Chapter 8 presents the numerical simulations for heat and mass transfer in supercritical carbon dioxide in an enclosure. One of the walls is made of naphthalene which dissolves in supercritical carbon dioxide.

A summary and conclusions for the current research and the recommendations for future work are discussed in Chapter 9.
CHAPTER 2
BACKGROUND AND LITERATURE REVIEW

2.1 Thermally Induced Acoustic Waves: Predictions and Measurements

When a compressible fluid is subject to a rapid temperature increase along a solid wall, the fluid in the immediate vicinity of the wall is heated by conduction, and tends to expand. The inertia of the unperturbed fluid constrains the sudden expansion due to the sudden energy input, which induces a pressure wave called a thermally induced acoustic wave [1,2,21]. The thermally induced acoustic wave travels approximately at the local speed of sound within the fluid, and is reflected from the other wall when impinging on it. The wave repeatedly traverses the length between both walls, and its amplitude eventually damps out due to the viscous and thermal losses within the fluid.

There has been a considerable amount of analytic, numerical and experimental work studying thermally induced acoustic waves [1,2,22-26]. The past studies lie in three categories: thermally induced acoustic waves in ideal gases, in liquids and in supercritical fluids.

The thermally induced acoustic waves exhibit quite different characteristics in supercritical fluids and liquids when compared to those in ideal gases. In ideal gases, the thermally induced pressure waves exhibit long tails following a peak with a sharp wave front [2,27,28]. However in liquids, the thermally induced acoustic wave comprises of a peak proceeded by an undershoot [22]
2.1.1 Studies on Thermally Induced Acoustic Waves in Ideal Gases

In 1899 Lord Rayleigh [29], starting from the Navier-Stokes equations for compressible fluids, derived an approximate solution in the form of a series for the generation of a pressure wave with a finite amplitude induced by the imposition of a high temperature plane in a gas.

Trilling [21] studied the sound fields induced in a real gas by boundary temperature variations. A one-dimensional model and a two-dimensional model were linearized, and then long-term asymptotic solutions in closed form were derived by the Laplace transform technique. He showed how thermally induced acoustic waves can be produced by heating the boundaries of real gas without any boundary motion, and illustrated how the pressure, temperature and vorticity modes of motion interact with each other.

Larkin [30] was the first to develop a finite difference solution to study the thermally induced acoustic waves, in a confined gas, generated by instantly heating one parallel plate with the other maintained at the initial temperature. He employed an explicit finite difference scheme for the continuity and momentum equations, but an implicit one for energy equation. His solution predicted the generation of a thermally induced acoustic wave traveling approximately at the local acoustic speed in helium. Additionally, he found that neglecting the convective and dispersive terms in the momentum equation was not permissible.

Kassoy [31] studied the response of a perfect gas confined in a slot to a monotonically varying temperature disturbance at the boundaries, employing a variety of
perturbation methods. Kassoy investigated this problem in two time scales: acoustic time scale and conduction time scale. On the acoustic time scale, the solutions were based on a thin expanding conduction boundary layer adjacent to the slot wall and an isentropic core in which a thermally induced acoustic wave propagates. On the conduction time scale, the governing equations were found to include nonlinear convection and pressure work terms. Additionally, a weak acoustic field was found to be propagating in the spatially anisotropic system.

Radhwan and Kassoy [32] investigated the behavior of a gas confined between infinite parallel planar walls subjected to significant heat addition. The solutions were developed in terms of asymptotic expansions, which are valid when the ratio of the acoustic time scale to the conduction time scale is small.

Ozoe et al. [33,34] numerically studied one- and two-dimensional thermoacoustic convection, due to rapid heating of one of the enclosing walls, in a confined region filled with a compressible fluid. The compressible Navier-Stokes equations with constant viscosity, constant conductivity and negligible viscous dissipation were non-dimensionalized, and numerically solved by a finite-difference method, with the convective terms approximated by a first-order upwind scheme, which is a flip-flop procedure depending on the sign of the velocity. The pressure waves were captured with substantial numerical diffusion in their numerical results. It is also shown that the fluctuating velocity generated by the pressure waves greatly enhance the rate of heat transfer over that for pure transient conduction. Ozoe
et al. also studied effects of various parameters, such as gravity, viscosity, wall temperature, and fluid properties on the development of convection.

The description of thermally induced acoustic waves in a fluid by the Navier-Stokes equations (continuity equation, momentum equation and energy equation) along with an appropriate equation of state was compared, by Churchill and Brown [35], with that provided by the hyperbolic conduction equation. Churchill and Brown found that the Navier-Stokes equations with the equation of state provided a satisfactory model for thermally induce acoustic waves for short, long and intermediate times. However the simplified hyperbolic conduction equation model just provided a crude approximation, due to the postulate of a wave velocity with a constant magnitude, was thereby invalid for the wave generation, propagation and dissipation.

Brown and Churchill [24] also numerically investigated, with finite-difference method, thermally induced acoustic waves in a gas by rapid heating of a bounding solid surface. The numerical predictions showed that rapid heating of a solid surface bounding a region of gas generates a slightly supersonic wave with positive amplitude in pressure, temperature, density and mass velocity, which are in good qualitative agreement with prior experimental measurements.

Thermally induced acoustic waves in a semi-infinite medium [27] and in a confined medium [28] were theoretically and numerically investigated by Huang and Bau. Huang and Bau solved the linearized wave model with asymptotic methods and a numerically inverted
Laplace transform. Additionally, they solved the nonlinear Navier-Stokes equations by using two numerical algorithms: a finite difference, implicit, Crank-Nicolson scheme modified with Galerkin finite element interpolation in space [36] and a second-order accurate, general purpose, multi-dimensional solver for differential equations PDE2D [37]. Comparison between the linear and the nonlinear solutions demonstrated that the nonlinear waves damp and propagate a bit faster than their linear counterparts and that the linear approximation is adequate in many practical situations.

Farouk et al. [2] used a control-volume-based flux-corrected transport algorithm to predict the early time behavior of thermally induced acoustic waves in a compressible-fluid filled cavity. In their numerical model, the temperature dependent fluid properties were used. Aktas and Farouk [38] recently studied the effect of gravity on the fluid motion generated by the thermally induced acoustic waves in a rectangular enclosure. The gravitational acceleration was found to have a negligible effect on the behavior of thermal induced acoustic wave for early times. A uniformly heated side wall was considered and the flow development of thermally induced acoustic waves under zero-gravity conditions was not studied.

Lin et al. [39] numerically investigated the flows generated by thermally induced acoustic waves in an enclosure with and without gravity, using a control-volume-based flux-corrected transport algorithm. Lin et al. considered that the left wall of the enclosure was heated rapidly either in a spatially uniform or a nonuniform manner, whereas the right wall was held at the initial temperature of the gas. Their simulations showed that, at zero gravity,
the spatially nonuniform heating induced a vortical flow in the enclosure, similar to that found in buoyancy-induced flows in a side-heated enclosure. Lin et al. [40] also numerically studied the interactions of the thermally acoustic waves produced by rapid heating of a wall with the buoyancy-induced flows in air filled rectangular enclosures.

Experimental measurements of pressure waves generated by rapid heating of a surface were reported by Brown and Churchill [1,41]. In their experiments, the rapid heating procedure was achieved by an R-C circuit. These pressure measurements in the wall of the closed chamber clearly demonstrated the generation of acoustic waves by rapid heating of a wall.

Lin and Farouk [42] experimentally investigated the behaviors of thermally induced acoustic waves generated by rapid heating of a bounding solid wall in a closed cylindrical tube. Lin and Farouk used a resistance-capacitance circuit to generate a rapid temperature Increase in a thin nickel foil located at one end of the closed cylindrical tube, generating a thermally induced acoustic wave in it. They measured the characteristics of generation, propagation, and decay of the acoustic waves with two different pressure transducers, and found that the measured decay rate of the acoustic waves was somewhat faster than the numerically predictions.

2.1.2 Studies on Thermally Induced Acoustic Waves in Liquids

Aktas et al. [22] are the only group who numerically studied the generation and propagation of thermally induced acoustic waves in liquids (water). Aktas et al. considered a
square enclosure filled with water as the computational domain. They calculated the thermodynamic properties of water via an accurate equation of state. They also studied the effect of the rapidity of the heating process on the production of thermally induced acoustic waves: impulse (sudden) heating and gradual heating of the left wall. Their simulations showed that thermally induced acoustic waves in liquids comprise of a peak proceeded by an undershoot, other than a long tail following a sharp wave front in ideal gases. Aktas et al. also found that thermally induced acoustic waves in water were significantly stronger for impulse heating than for gradual heating.

2.1.3 Studies on Thermally Induced Acoustic Waves in Supercritical fluids

A fluid is supercritical when the temperature and pressure are both above its critical temperature and critical pressure. Supercritical fluids exhibit many interesting characteristics, such as large compressibility, high density, large thermal conductivity, large specific heat, low sound speed, low viscosity and low thermal diffusivity. Thus supercritical fluids can be regarded as intermediate states between gases and liquids. Thermally induced acoustic waves are much stronger in supercritical fluids than in ideal gases under the same temperature increase and the rate of increase [25,26,43,44].

Because of the specific properties of supercritical fluids, such as the vanishingly small thermal diffusivity near the critical point, thermally induced acoustic waves play a dominant role in temperature equilibration [45,46]. Due to the vanishingly small thermal diffusivity near the critical point, very long time to reach equilibrium was expected for heat transfer in near-critical fluids. However, in both gravity and microgravity experiments the
temperature equilibration was found to occur rapidly. Under gravity, the fast time to reach temperature equilibrium was generally explained with the strong buoyancy convection in the fluids. Experiments by Staub in the TEXUS 8 sounding rocket seemed to indicate that the thermal equilibrium could be much faster than expected under microgravity [47,48]. In 1990 an experiment was carried out under microgravity in the TEXUS 25 sounding rocket [49]. In the experiment, the thermal boundary layer in the near-critical carbon dioxide expanded like a piston, due to the strong compressibility near the critical point, compressing the bulk fluid adiabatically. Therefore, the temperature rises in the fluid in a fast and homogeneous way. This mode of heat transfer has been called the piston effect [50,51].

2.2 Heat Transfer in Supercritical Fluids

During 1980s and 1990s, several teams in Europe found an interesting phenomenon in heat transfer in pure supercritical fluids[47,48]. The experimental study of the critical properties of a pure fluid near its liquid-vapor critical point is generally hampered by the occurrence of convection. This difficulty was considered to be overcome by carrying out the experiment in a microgravity environment [7,49]. Then the time required for a fluid sample to come into thermal equilibrium became an important consideration, because very long time was expected for heat transfer in near-critical fluids due to the vanishing thermal diffusivity near the critical point. However, in the experiments under microgravity[49], the temperature equilibration was found to occur rapidly. This interesting phenomenon led to the focus of many researchers on heat transfer in supercritical fluids.
Onuki et al. [7] theoretically studied the underlying physical mechanism. They assumed that a quantity of heat is introduced into a small subvolume $V_1$ of a supercritical fluid near its critical point, whose total volume is $V_1 + V_2$, where $V_2$ is the volume of the main bulk of the fluid. Onuki et al. [7] studied two cases for this problem: the fluid is kept isobaric or adiabatic. They found that for the adiabatic case the temperature of the main bulk of the fluid ($V_2$), which is untouched by any entropy transfer, shares to a considerable degree the temperature change with the small sub-region $V_1$, to which the actual heat transfer is confined. Through this process, the approach to equilibrium is thereby considerably facilitated. Then Onuki et al. pointed out that the very long heat relaxation for a supercritical fluid near its critical points exists only as an isobaric process, while for a clamped supercritical fluid kept at constant volume, the heat relaxation is much more rapid.

Boukari et al. [8] numerically studied one dimensional heat transfer in supercritical xenon under microgravity. The boundary of the supercritical xenon was quenched form 20 mK above $T_c$ (critical temperature) to 10 mK above $T_c$. Boukari et al. assumed that there was no velocity field within the xenon because of microgravity. Instead of usual Navier-stokes energy equation or usual conduction equation, Boukari et al. developed a new equation for conservation of energy:

Boukari et al. reported that the short equilibration time, which differed greatly from the relaxation time calculated from heat conduction along, can be accounted for by the adiabatic effect, and that temperature changes at the supercritical fluid boundary are rapidly
communicated to the interior of the fluid through adiabatic pressure changes, which travel by means of sound waves, thus the temperature can change very rapidly.

Figure 2.1 Temperature profiles for short times. The numbers refer to ratio $t/t_a$, with $t_a = 11.5\mu$s [6].

Zappoli et al. [6] studied the response to a boundary heating of supercritical CO$_2$ under microgravity by numerically solving the full one-dimensional Navier-Stokes equations. The equation of state for supercritical CO$_2$ used was the van der Waals equation. In their energy equation, the internal energy of supercritical CO$_2$ is supposed to by a function of temperature only, which is correct only for ideal gas. The PISO (Pressure Implicit with Splitting of Operators) method was employed to solve the one-dimensional Navier-Stokes equations. The temperature of one boundary was increased linearly by 13.0 mK in 1.3 ms. The temperature of the other boundary is kept constant, while Boukari et al. [8] employed an adiabatic boundary condition, because the adiabatic effect was considered crucial for the rapid thermal relaxation in supercritical fluids. Figure 2.1 shows the temperature profiles for short times. Figure 2.2 shows temperature increase ($\delta T = T - T_0$) for longer times. Both figures show rapid thermal equilibration in the supercritical CO$_2$. Zappoli et al. concluded that the
thermal energy is transformed into kinetic energy in a hot expanding boundary layer, which in turn transformed into thermal energy in the bulk fluid.

Figure 2.2 Temperature increase ($\delta T = T - T_i$) for longer times. The numbers refer to ratio $t/td$, with $td=157s$ [6].

Klein et al. [52] published their experimental results of temperature propagation in near-critical sulfur hexafluoride ($\text{SF}_6$). The $\text{SF}_6$ sample of critical density was housed in a cylindrical copper cell. The sample was rapidly cooled down from a temperature some milli-Kelvins above the critical temperature to a temperature some milli-Kelvins below the critical temperature under reduced gravity in a sounding rocket of the TEXUS (Technologische Experiment unter Schwerelosigkeit) program. At the critical temperature, thermal diffusivity of $\text{SF}_6$ is of the order of some $10^{-7}$ cm$^2$/s. This, along with the thermal length of 2 mm of Klein’s sample cell, leads to a thermal relaxation time of the order of about 100 h. However in Klein’s microgravity experiments, temperature changes were found propagating through the sample cell within seconds. Based on their experiment results and the theory of Onuki et al.[7], Klein et al. pointed out that using thermal diffusivity in relaxation time estimation only qualifies for samples at constant pressure. If the sample volume is fixed and gravity
convection is absent, the propagation of temperature changes into the interior region of the fluid near its critical point is mainly proceeding by adiabatic heating, like the temperature rise due to adiabatic compression. Temperature changes somewhere in the fluid cause expansion or contraction of the adjacent fluid. This in turn causes a pressure change throughout the sample volume with the consequence that the sample temperature is changed adiabatically and quickly.

Guenoun et al. [49] experimentally investigated the heat transport in near-critical carbon dioxide under microgravity. Their experimental system consisted of a transparent cell filled with carbon dioxide at critical density. The cell was mounted in a high precision thermostat, which can be cooled and heated rapidly. The internal volume of the experimental cell was a cylinder with a 10.106 mm diameter and a 7.48 mm thickness. The internal cylinder had two sapphire windows at the top and the bottom. A gold thread (0.3 mm diameter), which was thermally decoupled from the cell, was set in the center of the experimental cell. Measurements of temperature and density gradients were enabled by light transmissivity measurements through the cell sapphire windows. Figure 2.3 shows the sketch of Guenoun’s experimental cell.

Figure 2.3 Sketch of experimental cell, Guenoun et al. [49]
Guenoun et al. carried out their experiment on a 6-min flight of a sounding rocket. The system was kept at a constant temperature ($T_c + 2.3$ mK) at the beginning of the experiment. A thermal quench of 1 mK then was performed on the system. Guenon et al. observed homogeneous thermalization, with a time of thermalization less than 20 s. The typical diffusion time over distance $d$ (cell half-thickness) was defined as: $t_D = (D_T)^{-1}d^2$. $D_T$ is the thermal diffusivity. The typical diffusion time for CO$_2$ at this temperature is more than one week. The thermalization time thus is much smaller than the thermal diffusion time because of the so-called piston effect [49].

Bonetti et al. [53] experimentally studied the heat transport in supercritical carbon dioxide in the absence of gravity in a similar way as Guenoun et al. [49]. Bonetti’s experimental cell was a cylinder with an inner diameter of 11.6 mm and a thickness of 6.8 mm. Two sapphire windows were mounted at the top and the bottom of the cylinder. The cell was set in the center of a high precision thermostat. Two identical thermistors with nearly spherical shape were set in the fluid in the cell at an equal distance from the sapphire windows. Only one thermistor (referenced herein as TH1) was used to locally heat the fluid during experiment. TH1 measured the temperature of the thermal boundary layer, while the other thermistor measured the temperature in the bulk fluid. Interferometry was performed through those two sapphire widows to measure the density inhomogeneity within the bulk fluid. Figure 2.4 shows the evolution of a thermal boundary layer around the heating thermistor.
Figure 2.4 Evolution of a thermal boundary layer around the heating thermistor. (a) Before heating. (b) Experiment under gravity. (c) Experiment under microgravity. (d) Experiment under microgravity after 2s [53].

At equilibrium with homogeneous density, the interference fringes are straight and parallel. Density inhomogeneity in the bulk or at the cell walls results in a refractive index variation and fringes are shifted and/or distorted. From figure 2.4, we can see that under gravity a convective plume appeared in the fluid which destroyed the thermal boundary layer around the heating thermistor. While under microgravity no convective plume was observed around the heating thermistor. Instead a thermal boundary layer with a similar shape as the heating thermistor developed and expended around it. In the bulk fluid under microgravity, the fringes pattern with constant spacing corresponded to a spatially uniform density, which meant a spatially uniform bulk temperature, because we had an adiabatic process.
Zappoli and Carles [46] numerically studied the thermo-acoustic nature of piston effect. The problem geometry was a 1-D slab-like container filled with a near-critical fluid, which was submitted to a given heat flux at the boundary.

Zappoli’s governing equations [46] were non-dimensional 1-D compressible Navier-Stokes equations, in which the internal energy of near-critical fluids was assumed to be a function of temperature only. This assumption does not qualify for supercritical fluids. For other properties in the governing equations, such as thermal conductivity and viscosity, Zappoli and Carles [46] used the formulation: 

\[ \frac{\lambda'}{\lambda_0} = 1 + \Lambda \left[ \left( T' - T_c \right) / T_c \right]^{-0.5} \]

where \( \lambda' \) is the dimensional property, \( \lambda_0 \) denotes the property of the ideal gas. T’ is temperature and \( T_c \) is the critical temperature. \( \Lambda \) is a constant. Along with the van der Waals equation of state, the 1-D compressible Navier-stokes equations were solved by the matched asymptotic expansion method and multiple-time-scale techniques. Figure 2.5 shows temperature profiles at \( t' = 0.25 \) s and \( t' = 0.5 \) s for a 0.5 s boundary heating ting at constant flux. For following results, the initial and boundary conditions were: supercritical \( \text{CO}_2 \) initially at \( T_i = T_c + 1 \) K; left wall was heated by a 0.5 s long step heat flux; the geometry length was 12 mm. From figure 2.6, we can see the homogeneous heating of the bulk fluid. The homogeneous temperature increase in the bulk fluid was already very large at \( t' = 0.5 \) s. Figure 2.6 shows the magnified temperature profiles at different times.
Figure 2.5 Temperature profiles at $t' = 0.25$ s and $t' = 0.5$ s for a 0.5 s boundary heating at constant flux [46].

Figure 2.6 Magnified temperature profiles at different times [46].
It is interesting to note that Zappoli and Carles heated the left wall in three different cases. All the heat flux applied to the left wall just lasted for less than 2 s. The temporal variation of heat flux and bulk temperature for three different cases is shown in Figure 2.7. In the first case, a step heat flux was applied; in the second case, the heat flux increased and decreased linearly; in the third case, the heat flux increased linearly and then decreased exponentially. It can be seen in figure 2.7 that the evolution of the temperature was separated into two regimes. The first is a rapid increase by Piston Effect while the left wall was heated. The second is a slower decrease corresponding to the vanishing Piston Effect when the heat flux was stopped. In this paper, Zappoli and Carles numerically showed that the evolution of temperature in supercritical fluids was much faster than pure diffusion. The evolution of temperature was driven by a compression wave which was traveling at the acoustic speed back and forth in the fluid.

![Figure 2.7 Temporal variation of heat flux and bulk temperature for three different cases [46].](image-url)
Zappoli et al. [3] numerically studied the thermoacoustic and buoyancy-driven transport in a cavity filled with a near-critical fluid. Their problem is a two-dimensional cavity filled with near-critical CO$_2$ under normal gravity. The top, bottom and right walls of the cavity were insulated, while the temperature of the left wall was increased by 10 mK in 1.0 s. The length and height of the cavity are 1.0 cm. The initial temperature was homogeneous within the cavity, while the initial pressure and density were stratified due to gravity. The governing equations were non-dimensional 2-D compressible Navier-Stokes equations, in which the internal energy of near-critical fluids was assumed to be a function of temperature only.

For other properties, such as thermal conductivity and viscosity, Zappoli used this formulation: \( \frac{\lambda'}{\lambda'_0} = 1 + \Lambda \left[ \frac{(T' - T'_c)}{T'_c} \right]^{0.5} \), where \( \lambda' \) is the dimensional thermal conductivity, \( \lambda'_0 \) denotes the property of the ideal gas. \( T' \) is temperature and \( T_c \) is the critical temperature. \( \Lambda \) is a constant. The heat capacity at constant volume and viscosity was considered as constant in Zappoli’s computation. As for the equation of state for CO$_2$, Van der Waals equation was employed. The governing equations were numerically solved by means of a finite-volume method with SIMPLER algorithm, along with the acoustic filtering technique.

### 2.3 Heat and Mass Transfer in Supercritical Fluids

In recent years, supercritical carbon dioxide has been used widely in extraction and cleaning processes, as it is an excellent solvent. Supercritical fluids exhibit high compressibility while having high densities. This behavior leads to interesting heat transfer results influenced by generated acoustic waves when supercritical fluids are heated rapidly.
along an enclosure wall. Pitla et al. [54] developed a correlation to predict the heat transfer coefficient in supercritical carbon dioxide under forced cooling conditions based on the experimental data and numerical computations. Liao and Zhao [55] measured the average heat transfer coefficient for supercritical carbon dioxide flowing in horizontal mini/micro circular tubes under cooling condition. Liao and Zhao [56] also conducted experiments for convection of supercritical CO$_2$ in heated horizontal and vertical miniature tubes. Olson [57] measured the average heat transfer coefficients of supercritical CO$_2$ flowing in a horizontal tube with a diameter of 10.9 mm. Huai et al. [58] conducted experiments on convective heat transfer of supercritical CO$_2$ in mini channels, each with an inner diameter of 1.31 mm.

For supercritical carbon dioxide, Shan et al. [10] investigated the natural convection for a cold wall reactor. He et al. [59] carried out simulations of mixed convection heat transfer in carbon dioxide at supercritical pressure. An axisymmetric problem of upward flow in a heated vertical pipe was studied. Diffusion coefficient and solubility of naphthalene in supercritical carbon dioxide were determined by past studies [60-70]. Knaff and Schlunder [71] investigated mass transfer for dissolving solid naphthalene and caffeine in supercritical carbon dioxide. Sherwood numbers at the surface of naphthalene rod immersed in supercritical carbon dioxide were measured.

As supercritical fluids are more and more widely used in various industrial and laboratory processes, acoustically enhancement of transport in supercritical fluids begins to attract researching interest. Riera et al. [72] experimentally investigated mass transfer enhancement in supercritical fluids extraction by means of power ultrasound. Riera et al.
studied a supercritical carbon dioxide extraction of oil from particulate almonds. To examine the effect of the acoustic waves all experiments were performed with and without ultrasound.

### 2.4 Thermophysical Properties of Supercritical Fluids

Generally, the pressure a supercritical fluid is much higher than that when it can be treated as an ideal gas. Hence in supercritical region the p-ρ-T behavior exhibits strong non-linearity, especially near the critical point. Therefore, ideal gas law is not applicable any more for supercritical fluids. Consequently, equations of state for real gases are needed for numerical simulations for supercritical fluids. Several equations have been proposed for this purpose: van der Waals Equation, Redlich-Kwong Equation, Beattie-Bridgeman Equation, Benedict-Webb-Rubin equation and virial equation (an equation of state of gases that has additional terms beyond that for an ideal gas, which account for the interactions between the molecules). Among them, Benedict-Webb-Rubin equation is usually the most accurate. Whereas, van der Waals Equation has a historical value in that it was one of the first attempts to model the behavior of real gases, although its accuracy is often inadequate, especially in supercritical region.

### 2.5 Supercritical Fluids Applications

#### 2.5.1 Supercritical fluid extraction

Supercritical fluid extraction is a process that used supercritical fluids to selectively extract substances from solid or liquid mixtures. High product quality can be accomplished by fine-tuning pressure and temperature conditions of the fluid [73]. The advantages of supercritical fluid extraction (compared with liquid extraction) are that it is relatively rapid
because of the low viscosities and high mass diffusivities associated with supercritical fluids. In addition, there is no surface tension in a supercritical fluid, because there is no liquid / gas phase boundary in supercritical fluids. The extraction can be selective to some extent by controlling the density of the medium and the extracted material is easily recovered by simply depressurizing, allowing the supercritical fluid to return to gas phase and evaporate leaving no or little solvent residues. Carbon dioxide is the most common supercritical solvent. It is used on a large scale for the decaffeination of green coffee beans, the extraction of hops for beer production [74], and the production of essential oils and pharmaceutical products from plants.

2.5.2 Supercritical water oxidation

Supercritical water oxidation (SCWO), sometimes referred to as Hydrothermal Oxidation (HTO), is a process that occurs in water at temperatures and pressures above a mixture's thermodynamic critical point. Under these conditions water becomes a fluid with unique properties that can be used to advantage in the destruction of hazardous wastes such as PCBs. The fluid has a density between that of water vapor and liquid at standard conditions, and exhibits high gas-like diffusion rates along with high liquid-like collision rates. In addition, solubility behavior is reversed so that chlorinated hydrocarbons become soluble in the water, allowing single-phase reaction of aqueous waste with a dissolved oxidizer. The reversed solubility also causes salts to precipitate out of solution, meaning they can be treated using conventional methods for solid-waste residuals. Efficient oxidation reactions occur at low temperature (400-650 °C) with reduced NOx production.
SCWO can be classified as green chemistry or as a clean technology. The elevated pressures and temperatures required for SCWO are routinely encountered in industrial applications such as petroleum refining and chemical synthesis.

2.5.3 Supercritical fluid chromatography

Chromatography is an analytical technique used for the separation of complex chemical mixtures into individual components. In Supercritical Fluid Chromatography (SFC), the sample is carried through a separating column by a supercritical fluid (typically carbon dioxide) where the mixture is divided into unique bands based on the amount of interaction between the individual analytes and the stationary phase in the column. As these bands leave the column their identities and quantities are determined by a detector [75]. Supercritical fluid chromatography (SFC) is a relatively recent chromatographic technique, having been commercially available since only about 1982. As a result there is a large amount of research currently underway both in SFC method development and in hardware development. What differentiates SFC from other chromatographic techniques (gas chromatography (GC) and high performance liquid chromatography (HPLC)) is the use of a supercritical fluid as the mobile phase. Supercritical fluid chromatography has several main advantages over other conventional chromatographic techniques (GC and HPLC). Compared with HPLC, SFC provides rapid separations without the use of organic solvents. With the desire for environmentally conscious technology, the use of organic chemicals as used in HPLC could be reduced with the use of SFC. Because SFC generally uses carbon dioxide collected as a byproduct of other chemical reactions or is collected directly from the atmosphere, it contributes no new chemicals to the environment. In addition, SFC separations can be done
faster than HPLC separations because the diffusion of solutes in supercritical fluids is about ten times greater than that in liquids (and about three times less than in gases). This results in a decrease in resistance to mass transfer in the column and allows for fast high resolution separations. Compared with GC, capillary SFC can provide high resolution chromatography at much lower temperatures. This allows fast analysis of thermolabile compounds.

2.3.5 Nanoscale Material Fabrication in Supercritical Fluids

Because of their high mass diffusivities, supercritical fluids are capable of penetrating into porous solid materials dissolving organic compounds and transferring an effective mount of materials into very small spaces [76]. Because of their low viscosities, supercritical fluids can be transferred in pipelines and pumped to high pressures requiring less mechanical energy than liquids and subcritical gases. The tunable solvation characteristic of supercritical fluids, due to the continuously alterable density of supercritical fluids, makes supercritical fluids different from conventional solvents for nanotechnology. Another important advantage of supercritical fluids extraction is rapid separation of solutes that can be easily achieved by decreasing pressure.

Porous materials have very attractive physicochemical properties based on their porous structures and surfaces characteristics [77]. They are widely used as absorbents, catalysts, capacitors, and sensors. Many studies have been carried out about manufacturing porous materials with the use of templates, in which the porous materials are replicated according to the templates. However accurate replication is only possible above millimeter scale in conventional liquid solvents, because the high viscosity and surface tension prevents
penetration into the narrow gaps [77]. Because of the high diffusivity, low viscosity, zero surface tension, and controllable solubility, supercritical fluids can overcome the limitation of the diffusivity and mass transfer of conventional liquids solvents and transfer an effective amount of materials into very small spaces [77].
3.1 Overview

In this chapter, mathematical models describing the physical problems, and appropriate numerical schemes, with corresponding boundary conditions will be discussed for simulating thermoacoustic waves and transport in supercritical fluids. In addition, due to the special properties of supercritical fluids, the mathematical methods for numerical simulations in supercritical fluids will also be addressed in this chapter. In particular, fluids near the critical point exhibit interesting property variations. Unlike ideal gases, the internal energy of supercritical fluids is a function of both temperature and density (or pressure).

3.2 Governing Equations

A supercritical fluid is a compressible fluid (actually highly compressible), thus it is essentially characterized by a pressure $p$, a density $\rho$, with the corresponding temperature $T$. Hence, in this study, the compressible form of the Navier-Stokes equations, along with an equation of state, are employed for simulating transport and flow fields in supercritical fluids. For mass transfer in supercritical fluid, a species conservation equation is also considered with appropriate boundary conditions.

3.2.1 Two-dimensional Navier-Stokes Equations in Cartesian Coordinates for Heat Transfer in Supercritical Fluids
Since supercritical fluids are Newtonian fluids, the compressible form of two-dimensional Navier-Stokes equations can be expressed as:

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

\[
\frac{\partial (\rho u)}{\partial t} + \frac{\partial (\rho u^2)}{\partial x} + \frac{\partial (\rho uv)}{\partial y} = -\frac{\partial p}{\partial x} + \frac{\partial \tau_{xx}}{\partial x} + \frac{\partial \tau_{xy}}{\partial y}
\]  
(3.2)

\[
\frac{\partial (\rho v)}{\partial t} + \frac{\partial (\rho uv)}{\partial x} + \frac{\partial (\rho v^2)}{\partial y} = -\frac{\partial p}{\partial y} + \frac{\partial \tau_{xy}}{\partial x} + \frac{\partial \tau_{yy}}{\partial y} - \rho g
\]  
(3.3)

\[
\frac{\partial (\rho e)}{\partial t} + \frac{\partial (\rho eu)}{\partial x} + \frac{\partial (\rho ev)}{\partial y} + \frac{\partial (pu)}{\partial x} + \frac{\partial (pv)}{\partial y} = -\frac{\partial q_x}{\partial x} - \frac{\partial q_y}{\partial y} + \phi
\]  
(3.4)

\[p = f(\rho, T)
\]  
(3.5)

where x and y are the Cartesian coordinates, t is time, \(\rho\) is density, u and v are the horizontal and the vertical velocity components respectively, \(e\) is the total energy, p is pressure, and \(q\) is the heat flux. Here \(e = i + (u^2 + v^2)/2\), and \(i\) is the specific internal energy. Equation 3.5 \(p = f(\rho, T)\) is the equation of state.

The components of the non-dimensional stress tensor \(\tau\) (in Equations 3.2 and 3.3) are:

\[
\tau_{xx} = \lambda \left( \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) + 2\mu \frac{\partial u}{\partial x} = \left( \mu' + \frac{4}{3} \mu \right) \frac{\partial u}{\partial x} + \left( \mu' - \frac{2}{3} \mu \right) \frac{\partial v}{\partial y}
\]  
(3.6)

\[
\tau_{xy} = \lambda \left( \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) + 2\mu \frac{\partial v}{\partial y} = \left( \mu' + \frac{4}{3} \mu \right) \frac{\partial v}{\partial y} + \left( \mu' - \frac{2}{3} \mu \right) \frac{\partial u}{\partial x}
\]  
(3.7)

\[
\tau_{yx} = \mu \frac{\partial u}{\partial y} + \mu \frac{\partial v}{\partial x}
\]  
(3.8)
where $\mu$ is the dynamic viscosity, $\lambda$ is the second coefficient of viscosity and $\mu' = \lambda + 2\mu/3$ is the bulk viscosity. The components of the heat-flux vector (in Equation 3.4) are written as following, where $k$ is thermal conductivity and $T$ is temperature:

\[ q_x = -k \frac{\partial T}{\partial x} \]  
\[ q_y = -k \frac{\partial T}{\partial y} \]  

The viscous dissipation term (in Equation 3.4) is written as:

\[ \phi = \frac{\partial (\tau_{xx}u + \tau_{xy}v)}{\partial x} + \frac{\partial (\tau_{xy}u + \tau_{yy}v)}{\partial y} \]  

3.2.2 Two-dimensional Navier-Stokes Equations in Cartesian Coordinates for Heat and Mass Transfer in Supercritical Fluids

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

\[ \frac{\partial (\rho u)}{\partial t} + \frac{\partial (\rho u^2)}{\partial x} + \frac{\partial (\rho uv)}{\partial y} = -\frac{\partial p}{\partial x} + \frac{\partial \tau_{xx}}{\partial x} + \frac{\partial \tau_{xy}}{\partial y} \]  

\[ \frac{\partial (\rho v)}{\partial t} + \frac{\partial (\rho uv)}{\partial x} + \frac{\partial (\rho v^2)}{\partial y} = -\frac{\partial p}{\partial y} + \frac{\partial \tau_{xy}}{\partial x} + \frac{\partial \tau_{yy}}{\partial y} - \rho g \]  

\[ \frac{\partial (\rho e)}{\partial t} + \frac{\partial (\rho eu)}{\partial x} + \frac{\partial (\rho ev)}{\partial y} + \frac{\partial (pu)}{\partial x} + \frac{\partial (pv)}{\partial y} = -\frac{\partial q_x}{\partial x} - \frac{\partial q_y}{\partial y} + \phi \]  

\[ \frac{\partial \rho_n}{\partial t} + \frac{\partial (\rho_n u)}{\partial x} + \frac{\partial (\rho_n v)}{\partial y} = -\frac{\partial J_x}{\partial x} - \frac{\partial J_y}{\partial y} \]  

\[ p = f(\rho, T) \]
where $x, y$ are the Cartesian coordinates, $t$ is time, $\rho$ is density of the solution $\rho = \rho_{co_2} + \rho_n$, Here $\rho_{co_2}, \rho_n$ are the partial densities of CO$_2$ and naphthalene respectively, $u$ and $v$ are the velocity components, $e$ is the total energy, $p$ is pressure, and $q$ is the heat flux. Here $e = i + 0.5 (u^2 + v^2)$, and $i$ is the internal energy of the solution.

The components of the non-dimensional stress tensor $\tau$ (in Equations 3.13 and 14) are:

$$
\tau_{xx} = \lambda \left( \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) + 2 \mu \frac{\partial u}{\partial x} = \left( \mu' + \frac{4}{3} \mu \right) \frac{\partial u}{\partial x} + \left( \mu' - \frac{2}{3} \mu \right) \frac{\partial v}{\partial y} \tag{3.18}
$$

$$
\tau_{xy} = \lambda \left( \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) + 2 \mu \frac{\partial v}{\partial y} = \left( \mu' + \frac{4}{3} \mu \right) \frac{\partial v}{\partial y} + \left( \mu' - \frac{2}{3} \mu \right) \frac{\partial u}{\partial x} \tag{3.19}
$$

$$
\tau_{yy} = \mu \frac{\partial u}{\partial y} + \mu \frac{\partial v}{\partial x} \tag{3.20}
$$

where $\mu$ is the dynamic viscosity, $\lambda$ is the 2nd coefficient of viscosity and $\mu' = \lambda + \frac{2}{3} \mu$ is the bulk viscosity. The components of the heat-flux vector are written as following, where $k$ is thermal conductivity, $T$ is temperature and $D$ is the diffusion coefficient.

$$
q_x = -k \frac{\partial T}{\partial x} \tag{3.21}
$$

$$
q_y = -k \frac{\partial T}{\partial y} \tag{3.22}
$$

$$
J_x = -\rho D \frac{\partial (\rho v / \rho)}{\partial x} \tag{3.23}
$$

$$
J_y = -\rho D \frac{\partial (\rho v / \rho)}{\partial y} \tag{3.24}
$$

The viscous dissipation term (in Equation 3.15) is written as:
\[
\phi = \frac{\partial (\tau_{xx} u + \tau_{xy} v)}{\partial x} + \frac{\partial (\tau_{xy} u + \tau_{yy} v)}{\partial y}
\]  
(3.25)

### 3.2.3 Boundary Conditions

No-slip boundary conditions are applied for all solid walls. In addition, a high-order non-dissipative algorithm such as FCT requires rigorous formulation of boundary conditions [78]. Otherwise, numerical solutions may show spurious wave reflections at the regions close to boundaries and nonphysical oscillations arising from instabilities. In the present computational method, the treatment proposed by Poinsot and Lele [79] was followed for implementing the boundary conditions for density. This method is based on the theory of wave characteristics and avoids incorrect extrapolations and overspecified boundary conditions.

Along any solid wall, the density is calculated from

\[
\left( \frac{\partial \rho}{\partial t} \right)_w + \frac{1}{c_w} \left( \partial \rho \partial n + \rho c \partial u_n \partial n \right)_w = 0
\]

where the subscript \( w \) signifies the location of the wall, and \( n \) is the direction normal to the wall. Following the procedure described by Poinsot and Lele, the density along a vibrating wall is calculated by:

\[
\frac{\partial \rho}{\partial t} = \frac{\rho \gamma}{c_w} \frac{\partial u_w}{\partial t} + \frac{\rho \gamma (u_w - c_w)}{c_w} \frac{\partial u}{\partial x} - \frac{\gamma (u_w - c_w)}{c_w^2} \frac{\partial \rho}{\partial x}
\]

(3.27)
3.3 Thermophysical Property Evaluations for Supercritical Fluids

Due to the specific properties of supercritical fluids, mathematical methods regarding them, such as equation of state and internal energy, are quite different from those for ideal gases. In this section, the difference between supercritical fluids and ideal gases in equation of state, internal energy, will be discussed. In addition, the mathematical techniques employed for representing and handling these specific properties will be addressed.

3.3.1 Equation of State

For ideal gases, the equation of state is \( p = \rho RT \), where, \( R \) is the specific gas constant. This is a linear equation of state, which makes the numerical simulations for ideal gases easier to handle and less time-consuming, because of the good characteristics of linear equations. While for supercritical fluids, the equation of state exhibits strong non-linearity, especially near the critical point.

Figure 3.1 shows the variation of pressure for carbon dioxide as a function of temperature and density, as an ideal gas. The variation of pressure for carbon dioxide as a function of temperature and density near the critical point is shown in Figure 3.2. Comparing Figures 3.1 and 3.2, we see that as an ideal gas (high temperature and low pressure), the pressure is a linear function of temperature and density, while in supercritical region (high pressure), the pressure is a strong non-linear function of temperature and density, especially near the critical point.
Figure 3.1 Variation of pressure for carbon dioxide as a function of temperature and density as an ideal gas

Figure 3.2 Variation of pressure for carbon dioxide as a function of temperature and density near the critical point [80]
3.3.2 Internal Energy

In addition to equation of state, other thermophysical properties show drastically different characteristics for supercritical fluids and ideal gases, especially internal energy.

Figure 3.3 Variation of internal energy for carbon dioxide as a function of temperature and density as an ideal gas

Figure 3.3 shows the variation of internal energy for carbon dioxide as a function of temperature and density as an ideal gas. As illustrated in figure 3.3, internal energy of an ideal gases is a function of temperature only. Internal energy of ideal gases is $i = C_v T$ (constant specific heat) or $i_a = i_b + \int_{T_b}^{T} C_v(T) dT$ (variable specific heat), where $T_0$ is the internal energy at the reference point. During the computation of direct numerical
simulations (DNS), internal energy of next time step is obtained after solving energy equation. Temperature of next time step needs to be retrieved from the internal energy. For ideal gases the process is quite direct and accurate: just solving \( i = C_v T \) (constant specific heat) or \( i_a = i_b + \int_{b}^{a} C_v dT \) (variable specific heat).

![Figure 3.4 Variation of internal energy for carbon dioxide as a function of temperature and pressure near the critical point [80]](image)

For a supercritical fluid, internal energy is not a function of temperature only anymore. The variation of internal energy for carbon dioxide as a function of temperature and pressure near the critical point is shown in Figure 3.4. As illustrate in Figure 3.4, internal energy of supercritical fluids is a function of both temperature and pressure, or a function of
both temperature and density. From thermodynamics, internal energy of a real gas is:

\[ i_a = i_b + \int_{T_b}^{T_a} C_v \, dT + \int_{v_b}^{v_a} \left[ T \left( \frac{\partial p}{\partial T} \right)_v - p \right] \, dv \]

### 3.3.3 Two Dimensional Least Squares Polynomial Fit.

As we discussed in previous two subsections, the equation of state and internal energy exhibit strong nonlinearity in supercritical region, especially in near-critical region. Other properties, such as acoustic speed, thermal conductivity and viscosity, follow the similar patterns as equation of state and internal energy. Thus constant values or linear equations are not valid for the representation of properties in the computation for supercritical fluids any more. Two dimensional polynomial fits are employed to represent equation of state, internal energy and other properties. The coefficients of two dimensional polynomial fits are obtained through least square method based on the data in the NIST Standard Reference Database 12 [80].

The general format of two dimensional polynomial fits is:

\[ p = \sum_{i=0}^{n_1} \sum_{j=0}^{n_2} c_{ij} T^i \rho^j \tag{3.28} \]

Where \( p \) is the pressure or other properties, \( T \) is temperature, \( \rho \) is density and \( c_{ij} \) is the coefficients of the polynomial fits. \( n_1 \) is the order of temperature and \( n_2 \) is the of density for the polynomial fits.
Least square method is employed to generate the coefficients of the polynomial fits based on the data in the NIST Standard Reference Database 12 [80]. Assuming the number of data points is \( m \), the summation of error squares is:

\[
\sum_{k=1}^{m} \left( \sum_{i=0}^{n1} \sum_{j=0}^{n2} c_{ij} T_k^i \rho_k^j - p_k \right)^2
\]  

(3.29)

In order to make the summation of error squares minimum, take the derivative of the summation with respect to the coefficient of \( c_{ab} \) and make it zero:

\[
\frac{\partial}{\partial c_{ab}} \left[ \sum_{k=1}^{m} \left( \sum_{i=0}^{n1} \sum_{j=0}^{n2} c_{ij} T_k^i \rho_k^j - p_k \right)^2 \right] = 0
\]

\[
\frac{\partial}{\partial c_{ab}} \left[ \sum_{k=1}^{m} \left( \sum_{i=0}^{n1} \sum_{j=0}^{n2} c_{ij} T_k^i \rho_k^j - p_k \right)^2 \right] = \sum_{k=1}^{m} \left\{ \frac{\partial}{\partial c_{ab}} \left[ \sum_{i=0}^{n1} \sum_{j=0}^{n2} c_{ij} T_k^i \rho_k^j - p_k \right]^2 \right\}
\]

\[
= \sum_{k=1}^{m} \left[ 2 \left( \sum_{i=0}^{n1} \sum_{j=0}^{n2} c_{ij} T_k^i \rho_k^j - p_k \right) T_k^a \rho_k^b \right] = 0
\]

Rewrite this equation, we get:

\[
\sum_{k=1}^{m} \left[ \left( \sum_{i=0}^{n1} \sum_{j=0}^{n2} c_{ij} T_k^i \rho_k^j - p_k \right) T_k^a \rho_k^b \right] = \sum_{k=1}^{m} \left[ \sum_{i=0}^{n1} \sum_{j=0}^{n2} c_{ij} T_k^{i+a} \rho_k^{j+b} - p_k T_k^a \rho_k^b \right] = 0
\]

\[
\sum_{k=1}^{m} \left( \sum_{i=0}^{n1} \sum_{j=0}^{n2} c_{ij} T_k^{i+a} \rho_k^{i+b} \right) = \sum_{k=1}^{m} p_k T_k^a \rho_k^b
\]  

(3.30)
Above equation is based on the derivative of the summation of error squares with respect to the coefficient of $c_{ab}$, where $a \in (0,1,2,\ldots,n_1)$ and $b \in (0,1,2,\ldots,n_2)$. After taking the derivative of the summation of error squares with respect to all coefficients, a set of linear equations is obtained. The number of equations is $(1 + n_1) \times (1 + n_2)$. The coefficients of the polynomial fits are obtained after solving these linear equations through Gauss method.

3.3.4 Retrieving Temperature from Internal Energy for Supercritical Fluids

In the direct numerical simulations in this study, internal energy for next timestep is obtained by solving the energy equation. Temperature for next timestep is then retrieved from internal energy. For an ideal gas, the process of calculating temperature as a function of internal energy is direct and accurate, because internal energy of an ideal gas is a function of temperature only. While for a supercritical fluid, the internal energy is a function of both temperature and density, and a polynomial fit is employed to calculate internal energy as a function of temperature and density. The process of retrieving temperature based on internal energy and density becomes complex and critical for the direct numerical simulations in supercritical fluids.

The first attempt to calculate temperature based on internal energy and density is to generate another polynomial fit for temperature as a function of internal energy and density. However huge errors will be brought during computations due to the inconsistency of the two polynomial fits for internal energy and for temperature:
\[ I = \sum_{i=0}^{n_1} \sum_{j=0}^{n_2} c_{ij} T^i \rho^j \]  
(3.31)

\[ T = \sum_{i=0}^{n_1} \sum_{j=0}^{n_2} c_{ij} I^i \rho^j \]  
(3.32)

Here \( I \) is the internal energy, \( T \) is temperature and \( \rho \) is density. In Equations 3.31 and 3.32, \( c_{ij} \) are the coefficients of the polynomial fits for internal energy and temperature, respectively. The coefficients of the polynomial fits, \( c_{ij} \), are different in different equations.

The final and successful method to retrieve temperature from internal energy and density is based on a fact that the order of temperature in the polynomial fit for internal energy as a function of temperature and density is second order. When the order of temperature \( n_1 = 2 \) (in Equation 3.40), the polynomial fit for internal energy as a function of temperature and density is accurate enough, with a higher order of density \( (n_2) \) in the polynomial fit.

With \( n_1 = 2 \), Equation 3.40 can be written as:

\[
I = \sum_{j=0}^{n_2} c_{0j} \rho^j + \sum_{j=0}^{n_2} c_{1j} T \rho^j + \sum_{j=0}^{n_2} c_{2j} T^2 \rho^j 
\]

\[
\left( \sum_{j=0}^{n_2} c_{2j} \rho^j \right) T^2 + \left( \sum_{j=0}^{n_2} c_{1j} \rho^j \right) T + \sum_{j=0}^{n_2} c_{0j} \rho^j - I = 0
\]  
(3.33)

Above equation can also be rewritten as:

\[ AT^2 + BT + C = 0 \]  
(3.34)
where \( A = \sum_{j=0}^{n^2} c_{2j} \rho^j \), \( B = \sum_{j=0}^{n^2} c_{1j} \rho^j \), \( C = \sum_{j=0}^{n^2} c_{0j} \rho^j - I \)

When retrieving temperature from internal energy and density, a quadratic equation (Equation 3.34) can be obtained based on the polynomial fit for internal energy as a function of temperature and density (Equation 3.40). There are two roots for the quadratic equation (Equation 3.31), one of which is negative and thus non-physical. The other root is positive, and thus the physical temperature. With this method, no error is introduced during retrieving temperature from internal energy and density.

### 3.4 Computational Model

The governing equations (except for the diffusion terms) are discretized via a finite-volume scheme with, LCPFCT, a flux-corrected transport (FCT) algorithm, while the diffusion terms (including the viscous term in the momentum equations and the conduction terms in the energy equation) are discretized through a second-order central difference scheme.

#### 3.3.1. The Flux-Corrected Transport (FCT) Algorithm

FCT is a kind of high-order, nonlinear, monotone, conservative, positivity preserving algorithms, which are accurate and resolve steep gradients [81]. LCPFCT is one of the latest one-dimensional Flux-Corrected Transport (FCT) algorithms with fourth-order accuracy and minimum residual diffusion [81]. It is written in Fortran, solving the general one-dimensional continuity equation with appropriate source terms.
LCPFCT itself can treat a one-dimensional convection equation with appropriate source terms in Cartesian, cylindrical or spherical, and generalized nozzle coordinates. A flexible set of boundary conditions for each equation can be selected by the appropriate choice of the arguments to the subroutine calls. LCPFCT can handle inflow, outflow, and reflecting wall conditions in several coordinates systems; there is also an option for periodic boundary conditions. Although LCPFCT only solves one-dimensional continuity equations, multidimensional problems may be solved by timestep-splitting techniques. Further details of the FCT algorithm used here are documented by Boris et al. [81].

LCPFCT implements an explicit solution of the general one-dimensional continuity equations:

$$\frac{\partial \Phi}{\partial t} + \frac{1}{r^{a-1}} \frac{\partial}{\partial r} \left( r^{a-1} \Phi v \right) + \frac{1}{r^{a-1}} \frac{\partial}{\partial r} \left( r^{a-1} D_1 \right) = C_2 \frac{\partial D_2}{\partial r} + D_3$$

(3.35)

where $v$ is the local convection velocity, $D_1$, $D_2$, and $D_3$ are additional source terms. The quantity $\Phi$ can be any flow variable, such as density $\rho$, momentum flux ($\rho u, \rho v, \rho w$) or energy $\rho e$. The value $C_2$ can either be a constant or a function of convected variables. Different one-dimensional geometries may be selected through variation of an input integer $\alpha$: where $\alpha=1$ is Cartesian or planar geometry, $\alpha=2$ is cylindrical geometry, and $\alpha=3$ is spherical geometry. By choosing $\alpha=4$ and writing problem-specific code for defining cell interface area and volume, we can define other useful coordinate system, such as elliptical coordinate or various nozzle geometry.
The application of FCT to this type of equation consists of three steps.

(1) Central difference the equation by volume integral:

Figure 3.5 shows a one-dimensional geometry in which the fluid is constrained to move along a tube. In the Lagrangian grid, the flux out of one cell on the interface can be calculated by

\[
\Delta v_{i+\frac{1}{2}} = v_{i+\frac{1}{2}}^f - v_{i+\frac{1}{2}}^g
\]

(3.36)

where the superscripts \( f \) and \( g \) indicate the velocities of the fluid and grid, respectively. By this definition, the explicit finite difference formula for the convection transport part of the convection equation is:

\[
\Lambda_i^* \Phi_i = \Lambda_i^o \Phi_i - \Delta t \cdot \Phi_i^o \frac{1}{2} A \Delta v_{i+\frac{1}{2}}^o + \Delta t \cdot \Phi_i^o \frac{1}{2} A \Delta v_{i+\frac{1}{2}}^o
\]

(3.37)

\( \Lambda \) is the cell volume, and \( A \) is the area of the interface. The superscripts \( o \) and \( * \) indicate the beginning and the end of the time step, respectively.

Figure 3.5 Geometry and layout of the LCPFCT finite volume grid
Add the source terms in Equation (3.37):

\[
\Lambda_i^o \Phi_{i} = \Lambda_i^o \Phi_{i}^o + \frac{1}{2} \Delta t \cdot A_{i+\frac{1}{2}} (D_{i+1} - D_{i}) - \frac{1}{2} \Delta t \cdot A_{i-\frac{1}{2}} (D_{i} + D_{i-1})
\]
\[
+ \frac{1}{4} \Delta t \cdot C_{2,i} \left( A_{i+\frac{1}{2}} + A_{i-\frac{1}{2}} \right) (D_{i+1} - D_{i-1}) + \Delta t \cdot \Lambda_i^o D_{3,i}
\]  (3.38)

(2) Add numerical diffusion to stabilize the algorithm:

\[
\Lambda_i^n \Phi_i = \Lambda_i^o \Phi_i + \nu \Lambda_{i+\frac{1}{2}} (\Phi_{i+1} - \Phi_i) - \nu \Lambda_{i-\frac{1}{2}} (\Phi_{i-1} - \Phi_i)
\]  (3.39)

\( \nu \) is the non-dimensional numerical diffusion coefficient, which appears as a consequence of considering adjacent grid points. Substituting Eqs. (3.36) and (3.37) into Eq. (2.39) with no source term gives

\[
\Lambda_i^n \Phi_i = \Lambda_i^o \Phi_i^o + \nu \Lambda_{i+\frac{1}{2}} (\Phi_{i+1} - \Phi_i) - \nu \Lambda_{i-\frac{1}{2}} (\Phi_{i-1} - \Phi_i)
\]  (3.40)

(3) Add antidiffusion to control the amount of numerical diffusion:

The antidiffusion is designed so that when the grid is the Langrangian case and the \( \Delta v_{i+\frac{1}{2}} \) vanishes in Eq. (2.40),

\[
\mathbf{f}^\text{ad}_{i+\frac{1}{2}} = \mu \Lambda_{i+\frac{1}{2}} (\Phi_{i+1} - \Phi_i)
\]  (3.41)

Applying Eq. (2.41) to Eq. (2.40) gives

\[
\Lambda_i^n \Phi_i = \Lambda_i^o \Phi_i^o + \left( \nu \Lambda_{i+\frac{1}{2}} - \mu \Lambda_{i+\frac{1}{2}} \right) (\Phi_{i+1} - \Phi_i) - \\
\left( \nu \Lambda_{i-\frac{1}{2}} - \mu \Lambda_{i-\frac{1}{2}} \right) (\Phi_i - \Phi_{i-1})
\]  (3.42)

The largest choice of the antidiffusion coefficient that still guarantees positivity linearly is
However, this antidiffusion coefficient is not large enough. To reduce the residual diffusion \((\nu - \mu)\) even further, the flux correction must be nonlinear, depending on the density gradient. This corrected antidiffusive flux is:

\[
 f^{c, i + \frac{1}{2}} = S_{i + \frac{1}{2}} \cdot \max \left\{ 0, \min \left[ S_{i + \frac{1}{2}}, \Lambda_i^n \left( \tilde{\rho}_{i+2} - \tilde{\rho}_{i+1} \right), \left| f^{ad, i + \frac{1}{2}} \right|, S_{i + \frac{1}{2}}, \Lambda_i^n \left( \tilde{\rho}_i - \tilde{\rho}_{i+1} \right) \right] \right\} \tag{3.44}
\]

Here, \(|S| = 1\) and \(\text{sign } S \equiv \text{sign}(\tilde{\Phi}_{i+1} - \tilde{\Phi}_i)\). The final value at the new time is

\[
 \Phi^n_i = \Phi_i - \frac{1}{\Lambda_i^n} \left( f^{c, i + \frac{1}{2}} - f^{c, i - \frac{1}{2}} \right) \tag{3.45}
\]

The value of the residual diffusion coefficient is observed as a critical parameter for the low Mach number and second order flow problems under investigation. The default value of the RDC is 0.999. RDC may be reset to unity for minimal residual diffusion or to slightly smaller values than 0.999.

The geometric variables used above in the Cartesian, cylindrical and spherical coordinates are defined as

\[
 \Lambda_i^n = \pi \left[ r_{i+\frac{1}{2}} - r_{i-\frac{1}{2}} \right] ^2 \quad \text{Cartesian} \tag{3.46}
\]

\[
 \Lambda_i^n = 4 \pi \left[ r_{i+\frac{1}{2}}^3 - r_{i-\frac{1}{2}}^3 \right] \quad \text{cylindrical}
\]

\[
 \Lambda_i^n = \frac{4}{3} \pi \left[ r_{i+\frac{1}{2}}^3 - r_{i-\frac{1}{2}}^3 \right] \quad \text{spherical}
\]

and
A_{i+\frac{1}{2}}^\frac{1}{2} = \pi [r_{i+\frac{1}{2}}^n + r_{i+\frac{1}{2}}^n] \quad \text{cylindrical} \quad (3.47)

\frac{4}{3} \pi [(r_{i+\frac{1}{2}}^n)^2 + r_{i+\frac{1}{2}}^n \cdot r_{i+\frac{1}{2}}^n + (r_{i+\frac{1}{2}}^n)^2] \quad \text{spherical}

### 3.3.2. Solving Procedure of LCPFCT

In the one-dimensional problem, the three conservation equations of gas dynamics are coupled, and need to be solved simultaneously.

\[
\frac{\partial \rho}{\partial t} = -\nabla \cdot (\rho \vec{v}) \quad (3.48)
\]

\[
\frac{\partial (\rho \vec{v})}{\partial t} = -\nabla \cdot (\rho \vec{v} \vec{v}) - \nabla p \quad (3.49)
\]

\[
\frac{\partial E}{\partial t} = -\nabla \cdot (E \vec{v}) - \nabla (\vec{v} p) \quad (3.50)
\]

Solving the coupled equations is done by first choosing an appropriate time step \( \Delta t \), then integrating from old time \( t^o \) to \( t^o + \frac{\Delta t}{2} \), and then integrate from time \( t^o \) to \( t^o + \Delta t \). The half time step integration is used to calculate time centered spatial derivatives and fluxes. The whole integration procedure for one time step is:

1. **Half time step integration:**
   a) Calculate \( \nu_i^o \) and \( p_i^o \) using the old known value of \( \rho_i^o \), \( \rho_i^o \nu_i^o \), \( E_i^o \) at the beginning of the time step;
   b) Convect \( \rho_i^o \) a half time step to \( \rho_i^{\frac{1}{2}} \);
c) Evaluate \(-\nabla p^o\) as the source term for momentum equation;

d) Convect \(p^o_i v^o_i\) a half time step to \(\frac{1}{2} \rho_i^2 v_i^2\);

e) Evaluate \(-\nabla (p^o v^o)\) as the source term for energy equation;

f) Convect \(E^o\) a half time step to \(\frac{1}{2} E_i^2\).

(2) Whole time step integration:

a) Calculate \(\frac{1}{2} v_i^2\) and \(\frac{1}{2} p_i^2\) using the old known value of \(\frac{1}{2} \rho_i^2, \frac{1}{2} \rho_i^2 v_i^2, \frac{1}{2} E_i^2\) at the half of the time step;

b) Convect \(p^o\) a full time step to \(p^1\);

c) Evaluate \(-\nabla \frac{1}{2} p^2\) as the source term for momentum equation;

d) Convect \(p^o_i v^o_i\) a full time step to \(p^1_i v^1_i\) using \(-\nabla \frac{1}{2} p^2\);

e) Evaluate \(-\nabla (p^1 v^2)\) as the source term for energy equation;

f) Convect \(E^o\) a full time step to \(E^1\).

(3) Repeat these two procedures above to do another time step from \(t^1\) to \(t^2\).

The above one-dimensional solving procedure can be used repeatedly to construct a multidimensional program by time step splitting in the different coordinate directions. The approach is straightforward. For example, in Cartesian coordinates, these equations can be
treated as the summary of three sets of one-dimensional equations in x-, y- and z- directions. Then the integration can be performed separately in the three directions.

To use this time step-splitting approach, the time step must be small enough that the distinct components of the flux do not change the cell-averaged values appreciably during the time step. This approach is second-order accurate as long as the time step is small, but there is still a bias built in depending on which direction is integrated first. To remove this bias, the sequence of directions of integration should be alternated.

3.3.3. Boundary Condition Configuration in LCPFCT

To implement boundary conditions in the numerical model, we develop extrapolations from the interior to ghost cells (see Figure 3.6) outside the computational domain that continue the mesh a distance beyond the domain boundary. The definition of the ghost cells allows the cells on the domain boundary to be treated as interior cells.

Figure 3.6 Ghost cell approach for a boundary at the cell interface
In this approach, the ghost cell value is given by

\[ \Phi^n_G = S^E_{EBC} \Phi^n_{IE} + V^E_{EBC} \]  \hspace{1cm} (3.51)

Here, the subscripts G and IE indicate the ghost cell and the first interior cell, respectively. The quantity \( S^E_{EBC} \) is the slope boundary factor for either boundary condition. The quantity \( V^E_{EBC} \) is specified as the value added to the ghost value.

(1) For a no-slip stationary wall

Momentum perpendicular to wall: \((\rho u_\perp)_G = -(\rho u_\perp)_{EI}\)

Momentum along wall: \((\rho u_\parallel)_G = -(\rho u_\parallel)_{EI}\)

Density: \(\rho_G = 2\rho_w - \rho_{EI}\)

Energy: \(E_G = 2E_w - E_{EI}\)

(2) For a no-slip vibrating wall

If the velocity of wall is only in the normal direction, then the only difference from the stationary wall is the equation of momentum perpendicular to the wall:

\((\rho u_\perp)_G = 2(\rho u_\perp)_w - (\rho u_\perp)_{EI}\)

(3) Symmetric line

Momentum perpendicular to boundary: \((\rho u_\perp)_G = -(\rho u_\perp)_{EI}\)

Momentum along boundary: \((\rho u_\parallel)_G = (\rho u_\parallel)_{EI}\)

Density: \(\rho_G = \rho_{EI}\)
Energy: \( E_G = E_{EI} \)

LCPFCT can also handle moving wall boundary conditions. In the numerical model (Figure 3.7), we define the dimensions of both cell center \( xcn(i) \) and cell boundary \( xbn(i) \). To deal with the computation with moving wall, we select several cells near the boundary as moving cells, while the other cells as fixed cells. The number of moving cell, \( i_{1-1} \), is selected so that the maximum grid size variation is less than 25%. This also means that \( xbn(i_1) \) is a fixed value. At every time step, we first update the wall location, \( xbn(1^a) \), according to the boundary condition. Then we can update the dimensions of every moving cells by,

\[
\begin{align*}
xcn(i) &= \frac{xbn(i_1)-xbn(i)}{(i_1-1) \times (2i-1)/2}, \quad i=1 \text{ to } i_{1} \\
xbn(i) &= xcn(i) + \frac{xbn(i_1)-xbn(i)}{(i_1-1)} \quad i=2 \text{ to } i_{1-1}.
\end{align*}
\]

Figure 3.7 Layout of the finite volume grid for moving wall
CHAPTER 4
NUMERICAL STUDY OF THERMALLY INDUCED ACOUSTIC WAVES IN A
SUPERCRITICAL FLUID LAYER

4.1 Introduction

The generation, propagation and dissipation of thermally induced acoustic waves in supercritical nitrogen and carbon dioxide are investigated via a high-order numerical scheme. A one-dimensional problem is considered where the supercritical fluid is contained between two parallel plates. The thermally induced acoustic waves are generated as the left wall is heated rapidly. The acoustic waves travel towards the opposing wall and get reflected and thereafter continue to reverberate between the two confining walls. The predicted temperature of the bulk supercritical fluid is found to increase homogeneously (so called piston effect).

In the present study, the generation and propagation of thermally induced acoustic waves in supercritical carbon dioxide are investigated via a high-order numerical scheme. The NIST Standard Reference Database 12 [80] is used to obtain the $\rho$-p-T relation for supercritical nitrogen and supercritical carbon dioxide as well as the internal energy $i (\rho, T)$, the acoustic speed $c (\rho, T)$, the thermal conductivity $k (\rho, T)$, and the viscosity $\mu (\rho, T)$ relations. The present results are compared directly with the past studies by Zappoli et al. [6] for supercritical carbon dioxide, and Nakano and Shiraishi [82] for supercritical nitrogen.
4.2 Problem formulation

A confined isothermal layer of supercritical fluid (at $T_1$, $p_i$) is considered. The left wall is heated under different heating rates to initiate the thermally induced acoustic waves (See Figure 4.1). The right wall is held at the initial temperatures. The thermally induced acoustic waves along with the flow and temperature fields are studied as functions of space and time. Both supercritical nitrogen ($p_i > p_{cr}; T_i > T_{cr} \text{ where } p_{cr} = 3.39 \text{ MPa and } T_{cr} = 126.2 \text{ K}$) and supercritical carbon dioxide ($p_i > p_{cr}; T_i > T_{cr} \text{ where } p_{cr} = 7.377 \text{ MPa and } T_{cr} = 304.128\text{K}$) are considered for the simulations.

![Figure 4.1 Schematic of the problem geometry](image)

4.2.1 Equation of state and thermodynamic properties of supercritical nitrogen and carbon dioxide

The equation of state describing the $p$-$\rho$-$T$ relation of supercritical fluids is complex, particularly in the near-critical region. It has been shown earlier [83] that the van der Waals equation does not represent the $p$-$\rho$-$T$ relation of supercritical nitrogen and carbon dioxide.
accurately near the critical point. In this study, we use the NIST Standard Reference Database 12 [80] for the evaluation of the thermodynamic properties of supercritical nitrogen and supercritical carbon dioxide. The NIST Standard Reference Database 12 [80] is based on accurate experimental data with sound scientific judgment. Along with the data for equations of state for thirty two fluids, it provides all kinds of thermodynamic and transport properties, such as: density, internal energy, enthalpy, Cv, Cp, sound speed, viscosity, thermal conductivity, Prandtl number etc. All the fluid property data in this study and thesis are obtained from the NIST Standard Reference Database 12 [80].

![Graphs showing variations of pressure](image)

Figure 4.2 Variations of pressure $p$ for (a) carbon dioxide and (b) for nitrogen as functions of temperature $T$ and density $\rho$ near the respective critical points [80]

Variations of pressure for carbon dioxide and nitrogen as functions of temperature and density near the respective critical points are shown in Figures 4.2(a) and 4.2(b) respectively, where the $p$-$\rho$-$T$ relations are found to be strongly nonlinear, especially near the
critical points. While the critical temperatures and pressures for carbon dioxide and nitrogen are different, the overall behaviors of the $p$-$\rho$-$T$ variations are quite similar for both fluids.

Figures 4.3(a) and 4.3(b) show the variations of thermal diffusivity for carbon dioxide and nitrogen as functions of temperature and pressure near the respective critical points. It is noted that thermal diffusivities are very small near the critical points. Figures 4.4(a) and 4.4(b) show the variations of internal energy ‘$i$’ for carbon dioxide and nitrogen as functions of temperature and pressure near the respective critical points. The internal energy values shown are on a relative scale, based on chosen reference values [80]. In most of the past studies [3,6,46,82,84,85], the internal energy of the supercritical fluid was considered to be a function of temperature only. The present results are based on the variations of the internal energy as shown in Figures 4.4(a) and 4.4(b).

![Figure 4.3 Variations of thermal diffusivity $\alpha$ for (a) carbon dioxide and (b) for nitrogen as functions of temperature and pressure near the critical points [80]](image)
Figure 4.4 Variations of internal energy \(i\) for (a) carbon dioxide and (b) for nitrogen as functions of temperature and pressure near the critical points [80]

The variations of thermal conductivity for carbon dioxide and nitrogen as functions of temperature and pressure near the respective critical points are shown in Figures 4.5(a) and 4.5(b). It is interesting to note that thermal diffusivities reach their maximums at the critical points, while the thermal diffusivities are at their minimum at the critical points. This is due to the high densities at the critical points. Figures 4.6(a) and 4.6(b) show the variations of viscosity for carbon dioxide and nitrogen as functions of temperature and pressure near the respective critical points. Just like for ideal gases, the viscosities for supercritical fluids keep decreasing as the temperature increases at a constant pressure. However, at the pseudo critical point for any pressure, the viscosity exhibits an inflexion.
Figure 4.5 Variations of thermal conductivity for (a) carbon dioxide and (b) for nitrogen as functions of temperature and pressure near the critical points [80]

Figure 4.6 Variations of viscosity for (a) carbon dioxide and (b) for nitrogen as functions of temperature and pressure near the critical points [80]
Figures 4.7(a) and 4.7(b) show the variations of acoustic speed for carbon dioxide and nitrogen as functions of temperature and pressure near the respective critical points. From Figures 4.3 and 4.7, we see that acoustic speeds follow the same variation patterns as thermal diffusivity for supercritical fluids. The absolute minimum appears at the critical point. Moreover, for every constant pressure, there a local minimum at the corresponding pseudo critical point.

![Figure 4.7 Variations of acoustic speed for (a) carbon dioxide and (b) for nitrogen as functions of temperature and pressure near the critical points][80]

In summary, there are three variation patterns for thermophysical properties of supercritical fluids: 1) The property absolute maximum appears at the critical point, and exhibits a local maximum at the pseudo critical point for every constant pressure. Moreover, the local maximum decreases with increasing pressure. 2) The property exhibits the absolute minimum at the critical point, and there is a local minimum at the pseudo critical point for
every constant pressure. And the local minimum increases as pressure increases. 3) The property keeps increasing or decreasing as temperature increases. Besides, an inflexion appears at the pseudo critical point for every constant pressure.

Polynomial fits are used in the present study to represent the $p(\rho, T)$, $i(\rho, T)$, $k(\rho, T)$ and $\mu(\rho, T)$ relations in the NIST Standard Reference Database 12 [80]. The forms of fitted equations are shown in equations (4.1) and (4.2) below for $p(\rho, T)$, and $k(\rho, T)$ only. Similar forms are also used for $i(\rho, T)$, and $\mu(\rho, T)$.

$$p(\rho, T) = \sum_{i=0}^{n} \sum_{j=0}^{m} a_{ij}(\rho - \rho_r)^i(T - T_r)^j$$

(4.1)

$$k(\rho, T) = \sum_{i=0}^{n_1} \sum_{j=0}^{m_1} b_{ij}(\rho - \rho_r)^i(T - T_r)^j$$

(4.2)

Equation (4.1) is the fitted equation for pressure as a function of density and temperature, where $a_{ij}$ are the fitted coefficients. $\rho_r$ and $T_r$ are the reference density and temperature respectively and $n$ and $m$ are the orders of the fitted equation. In equation (4.2), $b_{ij}$ are the fitted coefficients, $n_1$ and $m_1$ are the orders of the fitted equation. In the current study, a sixth order polynomial fit is used for the properties. The pressure deviation for the fitted data from the original data in the NIST Standard Reference Database 12 [80] ranges from 0.00002% to 0.01%. The viscosity deviation for the fitted data from the original data in the NIST Standard Reference Database 12 [80] ranges from 0.00001% to 0.001%, the thermal conductivity deviation ranges from 0.01% to 0.9%, and fitted internal energy deviation from the original data ranges from 0.0001% to 0.6%. 

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4.2.2 Numerical model

An explicit finite-volume approach is used to solve the discretized form of the governing equations. The convective terms are discretized using a flux-corrected transport (FCT) algorithm while the diffusion terms are discretized by a central-difference scheme [81]. The FCT approach for solving the convective transport terms gives higher-order accuracy and reduced numerical diffusion. Time-step splitting is used to couple the convection, diffusion and compressibility effects. The approach has been successfully used by Farouk et al. [2] to predict the generation and propagation of thermally induced acoustic waves in ideal gases and by Aktas et al. [22] in a liquid.

No-slip boundary conditions are used for velocity on both walls. The right wall temperature is maintained at the initial temperatures of the supercritical carbon dioxide. The wall boundary conditions for density are obtained using the formulation developed by Poinso t and Lele [79],

\[
\left. \frac{\partial \rho}{\partial t} \right|_w = \frac{1}{c} \left[ \frac{\partial \rho}{\partial n} \right]_w \mp \rho c \frac{\partial u}{\partial n} \right) \quad (4.3)
\]

Equation (4.3) was developed through a technique for specifying boundary conditions for hyperbolic systems, based on characteristic wave theory. This method has been extensively studied for Euler equations [86-88], and was extended to Navier-Stokes equations by Poinso t and Lele [79]. The mathematical background of boundary conditions based on characteristic wave analysis was discussed in the papers of Kreiss [86], and Engquist and Majda [88]. According to equation (12) the wall density is updated as a function of the density and velocity (parallel to the wall) gradients normal to the wall. Once
the density at the wall is computed from equation (12), the pressure is updated using the
equation of state for supercritical carbon dioxide as a function of wall temperature and wall
density. The above relation has been successfully used earlier for the prediction of thermally
induced acoustic waves in gases and water [89,90].

The stability of the explicit-time-marching solution algorithm is ensured by setting
the Courant number \(c\Delta t/\Delta x\) to be less than 1.0. Here \(\Delta t\) is the time step, \(\Delta x\) is the length of
the smallest computational grid and \(c\) is the local acoustic speed of the supercritical fluid at
the initial condition.

4.3 Results and discussion

4.3.1 Numerical simulations for thermally induced acoustic waves in ideal and high
pressure gases

Before studying thermally induced acoustic waves in supercritical fluids, we will
numerically investigate thermally induced acoustic wave shapes in ideal gases and high
pressure gases \((T_i = 310 \text{ K}, p_i = 0.1 \sim 7 \text{ MPa})\) in this section. This helps us to study the
different characteristic of thermally induced acoustic waves in intermediate stats between
ideal gases and supercritical fluids.

The temporal variation of the left wall temperature is given by Equation 4.9 in Section
4.3.5. All the information for the four cases in intermediate stats is listed in Table 4.1. The
computational domain \((L = 1.0 \text{ cm})\) is discretized uniformly with 300 grid. Time step is
determined by setting the courant number to be 0.4.
Table 4.1 Cases for thermally induced acoustic waves in ideal and high pressure gases

<table>
<thead>
<tr>
<th>Case #</th>
<th>L [cm]</th>
<th>Fluid</th>
<th>T_i [K]</th>
<th>p_i</th>
<th>T_{lr} [K]</th>
<th>T_r [K]</th>
<th>Bulk viscosity</th>
<th>A</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.1</td>
<td>1.0</td>
<td>CO_2</td>
<td>310</td>
<td>1 atm</td>
<td>320</td>
<td>310</td>
<td>zero</td>
<td>10</td>
</tr>
<tr>
<td>1.2</td>
<td>1.0</td>
<td>CO_2</td>
<td>310</td>
<td>1 atm</td>
<td>320</td>
<td>310</td>
<td>zero</td>
<td>1</td>
</tr>
<tr>
<td>1.3</td>
<td>1.0</td>
<td>CO_2</td>
<td>310</td>
<td>1 atm</td>
<td>320</td>
<td>310</td>
<td>zero</td>
<td>0.1</td>
</tr>
<tr>
<td>1.4</td>
<td>1.0</td>
<td>CO_2</td>
<td>310</td>
<td>1 atm</td>
<td>320</td>
<td>310</td>
<td>zero</td>
<td>0.01</td>
</tr>
<tr>
<td>1.5</td>
<td>1.0</td>
<td>CO_2</td>
<td>310</td>
<td>1 MPa</td>
<td>320</td>
<td>310</td>
<td>zero</td>
<td>0.01</td>
</tr>
<tr>
<td>1.6</td>
<td>1.0</td>
<td>CO_2</td>
<td>310</td>
<td>3 MPa</td>
<td>320</td>
<td>310</td>
<td>zero</td>
<td>0.01</td>
</tr>
<tr>
<td>1.7</td>
<td>1.0</td>
<td>CO_2</td>
<td>310</td>
<td>5 MPa</td>
<td>320</td>
<td>310</td>
<td>zero</td>
<td>0.01</td>
</tr>
<tr>
<td>1.8</td>
<td>1.0</td>
<td>CO_2</td>
<td>310</td>
<td>7 MPa</td>
<td>320</td>
<td>310</td>
<td>zero</td>
<td>0.01</td>
</tr>
</tbody>
</table>

In cases 1.1 ~ 1.4, the fluid is an ideal gas due to the low initial pressure (1 atm) and high initial temperature (310 K). The heating rates decreases gradually from case 1.1 to case 1.4. Cases 1.5 ~ 1.8 all exhibit the same heating rate (A = 0.01) and same initial temperature (310 K), while the initial pressure increases gradually from 1 MPa to 7 MPa. The initial temperature for cases 1.1 ~ 1.8 (310 K) is above the critical temperature of carbon dioxide (304.1 K), making sure that all cases are in the gas phase.

Figure 4.8 shows the temporal variations of pressure at the center point for cases 1.1 ~ 1.4. For the same initial state, faster heating rates generate stronger thermally induced acoustic waves and higher bulk fluid temperature rising rates. But the current numerical model does predict the same and correct acoustic speed for cases 1.1 ~ 1.4. Temporal variations of pressure at the center point for cases 1.1 ~ 1.4 are shown in Figure 4.9. As initial pressure increases, the thermally induced acoustic waves get stronger and stronger, and the acoustic speed also decreases with increasing initial pressure. When the initial state...
comes closer to the supercritical region, the long tail after the wave peak becomes shorter and shorter. In Section 4.3.5, we will see that the long tail will disappear in supercritical region.

Figure 4.8 Temporal variations of pressure at the center point for cases 1.1 ~ 1.4 (ideal carbon dioxide with different heating rates)

Figure 4.9 Temporal variations of pressure at the center point for cases 1.5 ~ 1.8
4.3.2 Comparison of Experimental Measurements and Numerical Simulations for thermally induced acoustic waves in ideal gases

Aktas and Farouk have successfully numerically simulated the generation and propagation of thermally induced acoustic waves in ideal gases (Nitrogen) [91] and also compared the numerical prediction with the experimental work by Brown and Churchill [1].

![Figure 4.10 Temporal variations of pressure at the center point for thermally induced acoustic waves in ideal nitrogen (numerical [91] and experimental [1] results)](image)

Figure 4.10 shows the Temporal variations of pressure at the center point for thermally induced acoustic waves in ideal nitrogen (numerical [91] and experimental [1] results). The numerical problem geometry [91] (a 40.7 cm long closed tube) and the heating condition are identical to those considered in the experimental study [1]. In the experiments [1], the heating was stopped when the maximum wall surface temperature was reached and the hot surface is subject to lose energy via conduction, convection and radiation during the experiments. The numerical computations [91] did not consider the cooling of the wall, and thus the wall surface temperature was assumed to be constant after the maximum was reached.

In Figure 4.10, qualitative match between the numerical and experimental results is observed. The numerical model [91] is able to predict the wave motion and the wave
reflections. Both numerical and experiment results show that thermally induced acoustic waves are moving with the local acoustic speed and the space-averaged pressure is increasing due to heat addition at the heated wall as the pressure wave amplitude decrease in the system.

Reviewing past work on experimental measurements and numerical simulations for thermally induced acoustic waves in air enable us to make following conclusions: 1) the numerical model, which utilizes LCPFCT and the wall boundary condition developed by Poinsot and Lele [79], is able to accurately predict the generation and propagation of thermally induced acoustic waves in ideal gases, especially at early times. 2) The shape of thermally induced acoustic waves in ideal gases predicted by the numerical model is physical, based on the comparison between the computational and experimental results. Based on the above conclusions, the same numerical model employed in current study is believed to be able to predict thermally induced acoustic waves in supercritical fluids with accurate equations of state and internal energy functions. Because the main differences between supercritical fluids and ideal gases are equations of state and internal energies.

4.3.3 Comparison with past studies for supercritical carbon dioxide [6]

The prediction of thermally induced acoustic waves with the present method is first compared with two past studies, as summarized in Table 4.2. In Case 1.9, we consider supercritical carbon dioxide as the medium [6], and supercritical nitrogen [82] in the other case, Case 1.10. The supercritical fluid confined within two parallel plates is initially considered to be quiescent for both cases. Thermally induced acoustic waves are generated by heating the left wall rapidly. In Case 1.9, the left wall temperature is linearly heated from 305.2 K to 305.213 K in 13 ms following [6], whereas in Case 1.10 (following [82]) the left
wall temperature is impulsively heated from 130.0 K to 131.0 K within the first time step only.

Table 4.2 Cases for comparison with past studies

<table>
<thead>
<tr>
<th></th>
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<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>1.9</td>
<td>0.25</td>
<td>CO₂</td>
<td>305.2</td>
<td>7.38</td>
<td>305.21/3</td>
<td>305.2</td>
<td>Zero</td>
<td>10 (linear heating)</td>
</tr>
<tr>
<td>1.10</td>
<td>1.0</td>
<td>N₂</td>
<td>130</td>
<td>3.815</td>
<td>131</td>
<td>130</td>
<td>Zero</td>
<td>2×10⁻⁷ (limited to the first time step)</td>
</tr>
</tbody>
</table>

Case 1.9 is simulated for comparison with results given by Zappoli et al. [6]. The computational domain length is 0.25 cm. A non-uniform mesh with 400 grid points, where \( Δx_{\text{max}}/Δx_{\text{min}} = 5.0 \), is employed. The time step is determined by setting the Courant number to be equal to 0.6. A linear heating rate (10 mK/ms) was considered in [10].

Figure 4.11 shows the spatial variations of pressure at different times for case 1.9 as obtained by Zappoli et al. [10] and by the present method. The numbers identifying each line in the figure refer to the ratio \( t/t_a \), where \( t_a = L/c \) is the acoustic time-scale; \( L \) is the distance between the two walls, and \( c \) is the acoustic speed at the initial conditions of the fluid. The acoustic speed used by Zappoli et al. [10] is \( c = 216 \) m/s while we obtain \( c = 187.4 \) m/s from the NIST Standard Reference Database 12 [80] for the same conditions. Two values of \( t/t_a \) are thus given in identifying the results in Figure 4.11, where the values within the parentheses correspond to the value \( c = 216 \) m/s used by Zappoli et al. [10].
A thermally induced acoustic wave is generated at the beginning of left wall heating process, and the wave travels between the left and right walls. For the present results, the arrows indicate the directions in which the wave is moving. Comparing results we see that somewhat higher pressure values are predicted by Zappoli et al. While the present results accurately predict the acoustic wave speed (as given by the NIST Standard Reference Database 12 [80]), \( c = 187.4 \text{ m/s} \), the acoustic speed is not easily determined from the results predicted by Zappoli et al. [6].

There is a fundamental difference between the model presented by Zappoli et al. and the present model. In the present study, the internal energy \( i \) is a function of temperature and density, and the equation of state provided by the NIST Standard Reference Database 12 [80] is employed, while in [10], the internal energy is considered to be a function of
temperature only, and the van de Walls equation is used as the equation of state. It is well known that van der Waals equation is quite inaccurate near the critical region. The NIST Standard Reference Database 12 [80] can provide accurate data for pressures up to 30 MPa and temperatures up to 523 K for carbon dioxide, with the estimated uncertainty ranging from 0.03% to 0.05% in density [80]. The initial density for case 1.9 predicted by the van der Walls equation is 276.2 kg/m$^3$, whereas it is 307.1 kg/m$^3$ in the NIST Standard Reference Database 12 [80].

![Image](image_url)

Figure 4.12 Spatial variations of $u$ component of velocity at different times for case 1.9.

Figure 4.12 shows the spatial variations of the $u$ component of velocity at different times for case 1.9 by the present method. The numbers refer to the ratio $t/t_a$, where $t_a = L/c$, as in Figure 4.11. Again, the numbers within the parentheses are based on $c = 216$ m/s, while the numbers outside are based on $c = 187.4$ m/s. During the heating process, the $u$ velocity varies spatially and continues to increase with time. The results in Figure 4.12 clearly show the acoustic wave front and how it is modified by each wall reflection.
Spatial variations of temperature at longer times (for Case 1.9) are shown in Figure 4.13. Following Zappoli et al., we have presented the results at times (non-dimensionalized by the diffusion time scale, $t_d = L^2/\alpha$, where $\alpha$ is the thermal diffusivity. In [10], $t_d = 157.0$ s, while we calculate $t_d = L^2/\alpha = 414.9$ s, based on $\alpha = 1.507 \times 10^{-8}$ m$^2$/s obtained from the NIST Standard Reference Database 12 [80]. The results given in Figure 4.13 contain the nondimensional time values by using both $t_d = 157.0$ s and 414.9 s where the values within the parentheses correspond to the value $t_d = 157.0$ s. Piston effect can be clearly observed in the results. As the thermal boundary layer is expanding toward the right wall, the temperature of the bulk fluid is heated homogeneously. Qualitatively speaking, the same results are also predicted by Zappoli et al. [6], although the piston effect shown in the current model is slightly weaker than that by Zappoli et al. [6].
In summary, the current model generates qualitatively similar results as the model developed by Zappoli et al. [6]. However, fundamental differences exist between the present results and those given by Zappoli et al. The generation and the subsequent motion of the acoustic waves are entirely absent in the results presented in [6]. Our present experimental results of thermally induced acoustic waves in supercritical carbon dioxide (see Chapter 6 of the present thesis) agree with our present predictions. The comparisons are discussed in detail in Chapter 6. However, two significant improvements are made in the present study: application of the equation of state from the NIST Standard Reference Database 12 [80] and consideration of internal energy ‘i’ as a function of temperature and density (see Figure 4.4). These improvements make the current model more complete, and enable it to correctly predict acoustic speeds of the thermally induced acoustic waves.

4.3.4 Comparison with a past study for supercritical nitrogen [82]

Case 1.10 (see Table 4.2) is simulated for comparison with the predictions by Nakano and Shiraishi [82] for supercritical nitrogen. In case 1.10, the computational domain length is 1.0 cm, the time step is $5.0 \times 10^{-8}$ s, and the mesh size is 401, same as those used in [82]. A rapid heating rate ($2 \times 10^7$ K/s) was considered in [18], however, the heating rate was limited to the first time step only. The same heating rate is followed in case 1.10.

Figure 4.12 shows the thermally induced pressure wave profiles predicted by the present method at early times for case 1.10. The outgoing wave profile is shown at 20 and 40 µs after the heating is initiated. Thereafter the wave is reflected from the right wall and moves back towards the left wall (the results at 60, 80 and 100 µs). It is noted here that the
heating rates and durations considered in [6] and [82] are drastically different, as reflected by the results shown in Figures 4.11 and 4.14.

Unlike the characteristic ‘long tails’ of the thermally induced acoustic pressure waves as found in ideal gases [2], the pressure wave signatures in Figure 4.14 show a near-symmetric profile at the beginning, due to the high density of the supercritical fluid and the extremely high heating rate used. The high density of the supercritical fluid causes the pressure rise to be damped out behind the wave, so a near-symmetric profile is generated in the pressure wave signature.

The present results (Figure 4.14) compare poorly with those given in [18] where the pressure profiles appear to be noisy and do not show any wave front. The pressure increases predicted by Nakano and Shiraishi are much lower than the values associated with the thermally induced acoustic waves shown in Figures 4.14. For the same problem geometry and heating conditions, a spatially uniform pressure rise predicted by Nakano and Shiraishi at 20 µs is just about 1.0 Pa, while the localized pressure rise (as shown in Figure 4.14) in the present predictions is about 9500.0 Pa. From the NIST Standard Reference Database 12 [80] property values, we find that \( \frac{\partial p}{\partial T} \bigg|_\rho \) is 11400.0 Pa/K at 3.815 MPa and 130.0 K for nitrogen (the initial condition for case 1.10). Hence for a 1.0 K increase in wall temperature, the predicted value of about 9500 Pa is much closer to the analytic value of 11,400 Pa. The cause of the discrepancy in [18] is most likely due to the approximate treatments of density boundary condition at the wall, the use of the van der Waals equation and the treatment of the
internal energy in [82]. As discussed earlier, we used the density boundary condition based on the wave theory, given by equation (11) [79]. As shown in Figure 4.4(a) earlier, internal energy is a function of temperature and density (or pressure) at the supercritical conditions and the treatment of internal energy as a function of temperature only (as followed in [82]) can lead to significant errors in the predictions.

Figure 4.14 Thermally induced pressure wave profiles at early times for case 1.10

Figure 4.15 shows the corresponding temperature wave profiles at the times reported in Figure 4.15. The corresponding results by Nakano and Shiraishi [82] (not shown) are starkly different from the crisp wave profiles shown in the present results. Instead of predicting wave shapes, the results by Nakano and Shiraishi show somewhat noisy near-uniform pressure and temperature rises, without showing any evidence of the characteristic thermally induced acoustic wave signature [28].
Figure 4.15 Thermally induced temperature wave profiles at early times for case 1.10

Figure 4.16 Temporal variations of temperature and pressure at the center of the computational domain for case 1.10

The temporal variations of the temperature and pressure at the center of the computational domain for case 1.10 are shown below in Figure 4.16. The intensity of the
thermally induced acoustic waves continuously decrease as the waves travel through the medium and get reflected from the two end walls. Both the temperature and pressure show gradual increase with time as the wave intensities diminish. The present results are similar to the temperature waveforms predicted and measured in our earlier studies of thermally induced acoustic wave generation and propagation in gases [42].

Figure 4.17 Spatial variations of temperature at t = 0.25 s, 0.5 s, 0.75 s and 1.0 s by the present method for case 1.10

The spatial variations of temperature at longer times (t = 0.25 s, 0.5 s, 0.75 s and 1.0 s) for case 1.2 are shown in Figure 4.17. To demonstrate the existence of the piston effect, we show the results on an expanded temperature scale. It is evident that the bulk fluid temperature increases homogeneously as the thermal boundaries are developing along the left and right walls.
Figure 4.18 shows the temporal variations of pressure and temperature at the center of the computational domain for case 1.10 for a time span of 0.0 – 5.0 s. The pressure and temperature at the center increase continuously. At this longer time scale, the fluctuations of temperature and pressure observed in Figure 4.16 (for early times) are not apparent in Figure 4.18. We note that the predicted thermally induced acoustic waves damp out quite rapidly for the conditions considered in case 1.10.

Comparison of the predicted temperature variations at longer times (5.0 s and 10.0 s) with those given in [82] are shown in Figure 4.19. The temperature profiles given in [82] compare poorly with the present predictions. The possible reasons for the discrepancies have already been discussed earlier (related to Figure 4.14). It is also conceivable that at these large times 5.0 s and 10.0 s, (compared to μs), the thermal diffusivity and viscosity will start affecting the results, as indicated in our results.
4.3.5 Parametric study of thermally induced acoustic waves in supercritical carbon dioxide

In this section we present results for thermally induced acoustic waves generated by a prescribed heating the left wall, which is realizable in experiments. The temporal variation of the left wall temperature is given by:

\[ T_l(t) = T_i + \Delta T \left[ 1 - \exp\left( -\frac{At}{\tau} \right) \right] \]  \hspace{1cm} (4.9)

where \( \tau = 2L/c \), L is the distance between the two plates, c is the acoustic speed at the initial conditions of the fluid and A is a constant that determines the heating rate. We define \( \Delta T = T_{l, f} - T_i \), where \( T_{l, f} \) (final temperature of the left wall) and \( T_i \) (initial temperature of the layer of fluid) are listed in Table 4.3 for different cases. For all cases, the right wall temperature \( T_r \)
= T_i. For faster heating rate, larger values are considered for the parameter 'A' in equation (13).

Table 4.3 Cases for parametric study of thermally induced acoustic waves

<table>
<thead>
<tr>
<th>Case #</th>
<th>L [cm]</th>
<th>Fluid</th>
<th>T_i [K]</th>
<th>p_i [MPa]</th>
<th>T_{lf} [K]</th>
<th>T_f [K]</th>
<th>Bulk viscosity</th>
<th>A</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.1</td>
<td>1.0</td>
<td>CO_2</td>
<td>310</td>
<td>7.9</td>
<td>312</td>
<td>310</td>
<td>zero</td>
<td>10</td>
</tr>
<tr>
<td>2.2</td>
<td></td>
<td>CO_2</td>
<td>310</td>
<td>7.9</td>
<td>312</td>
<td>310</td>
<td>zero</td>
<td>1</td>
</tr>
<tr>
<td>2.3</td>
<td></td>
<td>CO_2</td>
<td>310</td>
<td>7.9</td>
<td>312</td>
<td>310</td>
<td>zero</td>
<td>0.1</td>
</tr>
<tr>
<td>2.4</td>
<td></td>
<td>CO_2</td>
<td>310</td>
<td>7.9</td>
<td>312</td>
<td>310</td>
<td>zero</td>
<td>0.01</td>
</tr>
<tr>
<td>2.5</td>
<td></td>
<td>CO_2</td>
<td>310</td>
<td>7.9</td>
<td>312</td>
<td>310</td>
<td>zero</td>
<td>0.00</td>
</tr>
<tr>
<td>2.6</td>
<td></td>
<td>CO_2</td>
<td>304.14</td>
<td>7.385</td>
<td>304.145</td>
<td>304.14</td>
<td>zero</td>
<td>10</td>
</tr>
<tr>
<td>2.7</td>
<td></td>
<td>CO_2</td>
<td>304.14</td>
<td>7.385</td>
<td>304.145</td>
<td>304.14</td>
<td>non-zero</td>
<td>10</td>
</tr>
</tbody>
</table>

For the cases 2.1 – 2.7 listed in Table 4.3, we consider supercritical carbon dioxide as the medium. The computational domain (L = 1.0 cm) is discretized uniformly with 300 grid points for cases 2.1 – 2.5; and non-uniformly with 700 grid points for cases 2.6 and 2.7 where \( \Delta x_{\text{max}}/\Delta x_{\text{min}} = 5.0 \). Finer meshes are needed for cases 2.6 and 2.7 where the initial states are closer to the critical point (\( p_{\text{cr}} = 7.377 \) MPa and \( T_{\text{cr}} = 304.128 \)K). Time step of \( 6.8 \times 10^{-8} \) s are considered for all cases.

Figure 4.20 shows thermally induced pressure distributions at four widely separated times (\( t = 1.22 \times 10^{-4}, 1.14 \times 10^{3}, 5 \times 10^{-2} \) and \( 1.0 \times 10^{-1} \) s) for case 2.1. The rapid heating (\( A = 10.0 \) in Table 4.3) causes a pressure peak, which travels from the left end to the right as evident from the profiles at \( t = 1.22 \times 10^{-4} \) and \( 1.14 \times 10^{-3} \) s. The pressure wave generated along the left heated wall travels towards the right wall with a wave speed of about 197 m/s. This
predicted wave speed is close to the value (for acoustic speed) obtained from the NIST Standard Reference Database 12 [80] at the initial temperature and pressure of the supercritical carbon dioxide. Unlike the characteristic ‘long tails’ of the thermally induced acoustic pressure waves after the wave fronts in ideal gases [2], the pressure wave signatures in Figure 4.20 show a near-symmetric profile at the beginning \((t = 1.22 \times 10^{-4} \text{ s})\), due to the high density of the supercritical fluid. As the wave continues to bounce back and forth between the left and right walls, ‘a long tail’ eventually develops \((at \ t = 1.14 \times 10^{-3} \text{ s})\) and the wave shape becomes broader due to dissipation. As shown in Figure 4.20, at longer times \((t = 5 \times 10^{-2} \text{ and } 10^{-1} \text{ s})\), the wave peak is not noticeable; however, the pressure of the bulk fluid continues to increase (the piston effect).

![Figure 4.20 Thermally induced acoustic (pressure) wave profiles at four different times for case 2.1](image)

Spatial variations of the corresponding temperature wave at four different times for case 2.1 are shown in Figure 4.21. Comparing Figures 4.20 and 4.21, we see that temperature
and pressure profiles exhibit similar shapes, and the pressure and temperature waves travel at the same local sound speed. Thin thermal boundary layers start to form along the left and the right walls, which expand slowly toward the opposite walls with time.

![Figure 4.21 Spatial variations of temperature wave at four different times for case 2.1](image)

The temporal variation of pressure at the center of the domain is shown in Figure 4.22 at early times for case 2.1. Rapid temporal variation of the pressure is predicted as the thermally induced acoustic wave gets reflected from the walls repeatedly. As shown earlier in Figure 4.16 (for supercritical nitrogen), the amplitude of the pressure wave is progressively damped as the energy is dissipated in the media. Secondly, the pressure in the bulk fluid gradually increases as the wave travels back and forth and undergoes dissipative losses.
Figure 4.22 Temporal variation of pressure at the center point at early time for case 2.1

Figure 4.23 shows the temporal variations of pressure and temperature for case 2.1 at the center for longer time (up to 0.1 s). The rapid oscillations appear to die out by $t = 0.01$ s for both pressure and temperature. As the thermally induced acoustic wave progressively damps out, it heats up the bulk fluid homogenously, which drives the piston effect [3].
In Figures 4.22 and 4.23, we see that the temperature at the center of the computational domain is increasing, and the bulk fluid temperature also keeps rising homogeneously (see Figure 4.21, for example). This shows the existence of piston effect at times longer than the acoustic time-scale \( t/t_a \). The physics of piston effect in supercritical fluids is further discussed in the next sub-section.

The spatial temperature variation for case 2.1 at \( t = 0.1 \) s is shown in Figure 4.24. In the same figure we also show the spatial temperature variation obtained by only considering conduction in the medium. At \( t = 0.1 \) s, the thermal boundary layer expands fast toward the right wall, and the bulk fluid temperature has been increased homogeneously, which shows the existence of piston effect. Whereas for the conduction solution, the thermal boundary
layer expands slowly and the bulk temperatures remain essentially unchanged even after 0.1 s.

![Graph showing spatial temperature variations for case 2.1 and for conduction solution of case 2.1 at t = 0.1 s.](image)

**Figure 4.24** Spatial temperature variations for case 2.1 and for conduction solution of case 2.1 at t = 0.1 s

The effect of the wall heating rate on the thermoacoustic effect is presented next. We show the thermally induced acoustic (pressure) wave profiles for cases 2.1 (A = 10), 2.2 (A = 1), 2.3 (A = 0.1), 2.4 (A = 0.01) and 2.5 (A = 0.001) in Figure 4.25 at an early time. A lower ‘A’ value (Table 4.3) represents a slower heating rate, and consequently the corresponding result exhibits a weaker pressure wave as shown in Figure 4.25. For all cases (shown in Figure 4.25), the thermally induced acoustic waves are traveling towards the right wall after heating is initiated at the left wall. Except the amplitude values, the thermally induced acoustic (pressure) waves show the same wave shapes and the same wave speeds for the different heating rates.
4.3.6 Physical mechanisms in ‘piston effect’

Zappoli et al. [6] defined two time scales for characterizing the thermoacoustic problem: the acoustic time scale, \( t_a \) and the diffusion time scale, \( t_d \) both defined earlier. As can be seen in Figure 4.23 above, thermally induced acoustic waves gradually damp out. In the diffusion time-scale, the piston effect still continues to exist in supercritical fluids as shown in Figures 4.13, 4.17, and 4.21. We find from these results that the bulk fluid temperature increases homogenously with time. In Figures 4.20 and 4.21, we see that the pressure and the bulk fluid temperature are uniform at \( t = 0.1 \) s for case 2.1. Figure 4.26 below shows the spatial variation of velocity \( u \) at \( t = 0.1 \) s for Case 2.1. At this relatively large time, the thermally induced acoustic waves have been damped out and the velocity
profile exhibits extremely low values and a quasi-linear distribution through most of the bulk fluid.

![Graph showing spatial variation of velocity at t = 0.1 s for Case 2.1](image)

**Figure 4.26 Spatial variation of velocity at t = 0.1 s for Case 2.1**

Revisiting the energy equation (equation 3.4)

\[
\frac{\partial (\rho e)}{\partial t} + \frac{\partial}{\partial x} \left[ (\rho e + p) u \right] = \frac{\partial}{\partial x} \left[ \mu \tau \right] - \frac{\partial q}{\partial x}
\]

we note that the conduction and the viscous terms are negligible at t = 0.1 s for case 2.1. So in the bulk fluid \( \rho e \approx \rho i \) for low values of velocity. Variations of \( \rho i \) as a function of pressure and temperature for carbon dioxide for the conditions considered in case 2.1 are shown in Figure 4.27 as obtained from the NIST Standard Reference Database 12 [80]. It is interesting to note that the temperature increases with increasing pressure even for a constant \( \rho i \). This is an important source for the piston effect (so far not discussed in the literature) during the intermediate time (between the acoustic and the diffusion times). This effect is based on the
fact that the internal energy of a supercritical fluid is a function of temperature and density (or pressure) and decreases with increasing pressure for constant temperature.

Figure 4.27 Variation of $\rho_i$ as a function of pressure and temperature (NIST Standard Reference Database 12 [80]) for carbon dioxide for case 2.1

4.4 Conclusions

The generation and propagation of thermally induced acoustic waves in supercritical nitrogen and carbon dioxide are investigated numerically by solving a fully compressible form of the Navier-Stokes equations. The NIST Standard Reference Database 12 [80] is used to obtain the thermodynamic properties of supercritical nitrogen and carbon dioxide. Polynomial fits are generated to represent the equation of state and other thermodynamic property relations for supercritical nitrogen and carbon dioxide. The wave-theory based wall boundary condition for density [79], provides higher-order accuracy needed for accurate prediction of the thermally induced acoustic waves. The sources for piston effect in two time-scales are studied. In the acoustic time-scale, thermally induced acoustic waves play an
important role for piston effect. The cause of piston effect in the intermediate time scale is also explained by the fact that the internal energy of a supercritical fluid is a function of temperature and density (or pressure).

The details of generation, propagation and decay of thermally induced acoustic waves in supercritical fluids are presented for heating of a confining wall, under different heating rates. The model predictions for supercritical nitrogen and carbon dioxide are compared with recently published results by Zappoli et al. [6], and by Nakano and Shiraishi [82] respectively. While qualitative agreements are observed, the merits of the present method over the methods employed by Zappoli et al. and by Nakano and Shiraishi [82] are explained. Our experimental results (see Chapter 6) clearly indicate that the present computational results capture the details of the thermally induced waves more accurately than predicted in the past studies.
CHAPTER 5
NUMERICAL STUDY OF NATURAL CONVECTION IN SUPERCRITICAL FLUIDS IN AN ENCLOSURE

5.1 Introduction

Supercritical fluids are characterized by high densities, high thermal conductivities (compared to gases) and low viscosities, but low thermal diffusivities (compared to liquids). Due to the high compressibility, thermally induced acoustic waves are generated when supercritical fluids are heated/ cooled along any bounding surface. In this study, we obtain both short- and long-time solutions for convective flows in a supercritical carbon dioxide filled enclosure. The NIST Standard Reference Database 12 [80] is used to obtain the property relations for supercritical carbon dioxide. The generation and propagation of thermally induced acoustic waves produced immediately after rapid heating of a wall are investigated by solving the fully compressible Navier-Stokes equations with an accurate equation of state, via a high-order explicit numerical scheme. For longer time solutions, when the acoustic waves damp out, an implicit solution algorithm is used to simulate the heat transfer in the above enclosure filled with supercritical carbon dioxide for longer periods of time. This novel scheme allows us to investigate convective flows in an enclosure filled with supercritical fluid in a comprehensive manner.

5.2 Problem formulation

A supercritical carbon dioxide filled square enclosure with $L = H = 1.0$ cm is considered as shown in Figure 5.1. The left and right walls are isothermal at different
temperatures, while the top and bottom walls are kept insulated. The initial temperatures of the left and right vertical walls along with the supercritical carbon dioxide are at 310 K and the initial pressure in the enclosure is 7.9 MPa. It is noted that the critical pressure and temperature of carbon dioxide are $T_c = 304.1$ K and $p_c = 7.377$ MPa, respectively. The left wall is impulsively heated to 312 K, while the right wall is kept at the initial temperature 310 K.

![Figure 5.1 Computational domain for the two-dimensional enclosure](image)

### 5.2.1 Numerical scheme for short time solution

An explicit finite difference approach is used to solve the discretized form of the governing equations for the short time solutions. The convective terms are discretized using a flux-corrected transport (FCT) algorithm while the diffusion terms are discretized by a central-difference scheme. The FCT approach for solving the convective transport terms allows higher order accuracy and the ability to control numerical diffusion within the finite-difference grid. Time-step splitting approach is used to include the convection, diffusion and compressibility effects.
Employing a two-step predictor corrector scheme, the FCT algorithm ensures that all conserved quantities remain monotonic and positive. It first modifies the linear properties of a high order algorithm by adding diffusion during convective transport to prevent dispersive ripples from arising. This added diffusion is then removed in an anti-diffusion phase of the integration cycle. Hence, these calculations maintain the higher order accuracy without artificial viscosity to stabilize the algorithm. The present solution algorithm was successfully used by Farouk et al. [2,22] to predict the generation and propagation of thermally induced acoustic waves in ideal gases and in water.

No-slip boundary conditions are used for velocity on both walls. The right wall temperature is maintained at the initial temperatures of the supercritical carbon dioxide. The left wall temperature is given by Equation 17. The wall boundary conditions for density are obtained using the formulation developed by Poinsot and Lele [79], which is derived for the Euler equations and then generalized to the Navier-Stokes equations using characteristic wave relations.

\[
\frac{\partial \rho}{\partial t} + \frac{1}{c^2} \left( T + c \left[ \frac{\partial p}{\partial n_w} + \rho c \frac{\partial u}{\partial n_w} \right] \right) = 0
\]

(5.1)

The wall pressure is updated using the equation of state for supercritical carbon dioxide as a function of wall temperature and wall density.
The stability of the explicit-time-marching based solution algorithm was ensured by setting the Courant number \( (= c\Delta t/\Delta x) \) to 0.6. Here \( c \) is the acoustic speed, \( \Delta t \) is a typical time step and \( \Delta x \) is the length of the smallest computational grid.

5.2.2 Numerical scheme for long time solution

Due to the stability limit for explicit schemes, extremely small \( \Delta t \) must be employed for the FCT scheme, resulting in long computational time for long time solutions. Furthermore the thermally induced acoustic waves have been damped out in the long time simulations, thus FCT is not needed any more to capture the thermally induced pressure and temperature waves. A fully implicit scheme (CFD-ACE version 2007) is employed, which enable us to use large \( \Delta t \) to carry out long time simulations. Instead of the density-velocity formulation of the Navier-Stokes equations (as used for the explicit calculations), a pressure-velocity formulation of the Navier-Stokes equations are employed (for the implicit calculations). A relatively large time step \( \Delta t = 0.1 \text{ s} \) is employed for the simulations with CFD-ACE. The objective here was to obtain long-time solutions without the time-step restrictions (Courant condition) imposed in the explicit scheme.

For the implicit calculations, the fluid properties of supercritical carbon dioxide were also obtained from the NIST Standard Reference Database 12 [80]. Polynomial fits were generated to represent density, viscosity, specific heat, enthalpy and thermal conductivity as functions of temperature and pressure.
5.3 Results and discussions

Numerical simulations for heat transfer in supercritical carbon dioxide are performed in a square enclosure filled with supercritical carbon dioxide. For short-time solutions (employing an explicit time marching scheme), non-uniform meshes are employed. The non-uniform mesh is characterized by $\Delta y_{\text{ymax}}/\Delta y_{\text{ymin}} = 5$ and $\Delta x_{\text{xmax}}/\Delta x_{\text{xmin}} = 5$. For the long time solution (employing an implicit time-marching scheme), the square enclosure is discretized by a uniform mesh.

5.3.1 Short time solution

In this section we present the short-time-solution results for natural convection in supercritical carbon dioxide. The temporal variation of left wall

$$T_l(t) = T_i + \Delta T \left[ 1 - \exp \left( -\frac{At}{\tau} \right) \right]$$  \hspace{1cm} (5.2)

Here $\tau = 2L/c$, $L$ is the distance between the two plates, $c$ is the acoustic speed at the initial conditions of the fluid and $A$ is a constant that determines the heating rate. We define $\Delta T = T_{l,f} - T_i$, where $T_{l,f}$ (final temperature of the left wall) and $T_i$ (initial temperature of the fluid) are listed in Table 5.1 for different cases. For all cases, the right wall temperature $T_r = T_i$. For faster heating rate, larger values are considered for the parameter 'A' in equation (5.2)

Four cases are simulated for natural convection in supercritical carbon dioxide with the short time solution. Within these cases, the parameter $A$ varies for faster and faster heating rate on left wall. There are also two initial states within these cases: $T_i = 310$ K and $p_i = 7.5$ MPa, $T_i = 305$ K and $p_i = 7.5$ MPa.
Table 5.1 Description of cases for short time solution

<table>
<thead>
<tr>
<th>Case #</th>
<th>$T_i$ [K]</th>
<th>$T_{lf}$ [K]</th>
<th>$T_r$ [K]</th>
<th>$p_i$ [MPa]</th>
<th>Gravity</th>
<th>Gr</th>
<th>A</th>
<th>Mesh size</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>310</td>
<td>312</td>
<td>310</td>
<td>$7.5 + \int_0^l \rho gdy$</td>
<td>1g</td>
<td>2.089×10^8</td>
<td>500</td>
<td>301×101</td>
</tr>
<tr>
<td>2</td>
<td>310</td>
<td>312</td>
<td>310</td>
<td>$7.5 + \int_0^l \rho gdy$</td>
<td>1g</td>
<td>2.089×10^8</td>
<td>50</td>
<td>301×101</td>
</tr>
<tr>
<td>3</td>
<td>305</td>
<td>305</td>
<td>307</td>
<td>$7.5 + \int_0^l \rho gdy$</td>
<td>1g</td>
<td>2.228×10^9</td>
<td>10</td>
<td>401×121</td>
</tr>
<tr>
<td>4</td>
<td>305</td>
<td>305</td>
<td>307</td>
<td>$7.5 + \int_0^l \rho gdy$</td>
<td>1g</td>
<td>2.228×10^9</td>
<td>1</td>
<td>401×121</td>
</tr>
</tbody>
</table>

Thermally induced acoustic waves are generated in the enclosure due to the temperature rise of the left wall, and travel between the left and right walls of the enclosure. Thermally induced acoustic waves enhance heat transfer and fluid flow in the enclosure, thus playing an important role in temperature field and flow filed, especially for short time solution.

The temporal variation of pressure at center point of the enclosure for case 1 ((a) early time (b) longer time) is shown in Figure 5.2, which proves the existence of strong thermally induced acoustic waves in the enclosure. Pressure at the enclosure center undergoes rapid temporal variations as the thermally induced acoustic wave arrives at the center point when travelling between the left and right walls, as illustrated in part (a) of Figure 5.5. Two observations can be made from part (b) of this figure. First, the amplitude of the pressure wave is progressively damped as the energy is dissipated in the media. Secondly, the pressure in the bulk fluid gradually increased as the wave travels back and forth, and undergoes
dissipative losses. Part (a) of Figure 5.5 also shows that the pressure wave travels with a speed of about 197 m/s. This predicted wave speed is very close to, 196.7 m/s, the value obtained from the NIST Standard Reference Database 12 [80].

Figure 5.2 Temporal variation of pressure at the center for case 1: (a) early time (b) longer time

Figure 5.3 shows the temporal variation of x-component of velocity at the center point of the enclosure for longer time. The thermally induced acoustic wave undergoes many reflections between the left and right walls within the enclosure. The dark region in this figure reflects the highly oscillatory nature of the flow field induced by the thermally induced acoustic wave. The x- velocity component decays with every reflection and becomes quite small by 0.32 s.
The velocity vectors for case 1 at two different times (0.2 s and 0.32 s) are shown in Figure 5.4. We see that the oscillatory flow induced by the thermally induced acoustic wave almost damps out at these times. A buoyancy induced recirculating flow pattern develops in the enclosure, due to the high Grashof number ($Gr = \frac{g\beta(T_{l,f} - T_r)L^3}{\nu^2} = 2.089 \times 10^8$). Such a huge Grashof number for a 2 K temperature difference ($T_{l,f} - T_r = 2K$) is because of the extremely small kinematic viscosity of supercritical carbon dioxide ($\nu_{(310K,7.9MPa)} = \nu_{(310K,0.1MPa)} = 0.0085$).
Figure 5.4 Velocity vectors for case 1 at two different times: (a) 0.20 s and (b) 0.32 s
Figure 5.5 Spatial variation of temperature at the horizontal midplane \((y = 0.5 \text{ cm})\) for case 1 at \(t = 0.32 \text{ s}\).

Figure 5.6 Temporal variations of the heat fluxes at left and right walls for case 1.
Figure 5.5 shows the spatial variation of temperature at the horizontal midplane \((y = 0.5 \text{ cm})\) for case 1 at \(t = 0.32 \text{ s}\). As the thermally induced acoustic wave traverses between the left and right walls, the thermal boundary layers form and the temperature of bulk fluid also increases homogeneously. This phenomenon is called piston, which is already discussed in Chapter 4.

Temporal variation of the heat fluxes at the left and right walls for case 1 are shown in Figure 5.6. As the temperature of bulk fluid increases, the heat flux at the left wall decreases quickly, while the heat flux at the right wall increases gradually. At thermal equilibrium conditions, the heat fluxes at the left and right walls are equal to each other. The dark region in the figure at early time reflects the highly oscillatory nature of the heat fluxes at the left and right walls, due to the thermally induced acoustic waves. As the thermally induced acoustic wave impinges on the left wall, the heat flux at the left wall undergoes a rapid upward oscillation, while the wave impinges on the right wall, the heat flux at the right wall undergoes a rapid downward oscillation. As the thermally induced acoustic wave damps out within the enclosure, the heat flux oscillations also disappear simultaneously.
Figure 5.7 Temperature contours for case 1 at $t = 0.32$ s (a) zoomed region: $0 < x < 0.0015$ m

(b) entire enclosure: $0 < x < 0.01$ m
The temperature contours for case 1 at $t = 0.32$ s is shown in Figure 5.7. Part (a) shows a zoomed region: $0 < x < 0.0015$ m and part (b) shows the entire enclosure. In part (b) the temperature field is uniform in the entire enclosure, except in the thermal boundary layer, just as the uniform bulk temperature profile shown in Figure 5.5. The thermal boundary layer thickness is not a constant along the $y$ direction. It increases in the direction of $y$ axis. This is because the temperature contours are skewed by the recirculation in the enclosure, due to the buoyancy induced natural convection.

Four cases with different initial stats and left wall heating rates are simulated in this study. Figure 5.8 shows the temporal variation of pressure at the center point for all the cases. In Figure 5.8, “$A$” is the parameter for controlling left wall heating rate (see Equation 5.2) and a larger value of “$A$” means a faster left wall heating rate. Cases 1 and 2 are for one same initial state (310 K and 7.9 MPa), and Cases 3 and 4 are for another same initial state (305 K and 7.5 MPa). In Figure 5.8, we see that a lower left wall heating rate generates a weaker thermally induced acoustic waves within the enclosure. Comparing cases 1 and 2 with cases 3 and 4 shows that the rates under which the center point pressure increases are different for different initial states, due to the different thermophysical properties.
Figure 5.8 Temporal variations of pressure at the center point for cases 1, 2, 3 and 4.

Figures 5.9, 5.10 and 5.11 show the velocity vectors for cases 2 (t = 0.32 s), 3 and 4 (t = 0.2 s). At these times, the oscillatory flows induced by the thermally induced acoustic wave almost damps out. Additionally, buoyancy induced recirculating flow fields are developing in the enclosure for all these cases. In Figures 5.9, 5.10 and 5.11, an interesting phenomenon is observed: for cases 3 and 4, the buoyancy induced recirculating flow fields are stronger than those for cases 1 and 2. This can be explained by the fact that the Grashof number for cases 3 and 4 is about one order of magnitude higher than that for cases 1 and 2. In Figures 5.9, 5.10 and 5.11, we also see that the left wall heating rates do not affect the flow field as much as the initial states, because the heating rates only control the strength of the thermally induced acoustic waves, which damp out before the buoyancy induced flow fields appear in the enclosure.
Figure 5.9 Velocity vectors for case 2

Figure 5.10 Velocity vectors for cases 3
The temporal variations of the heat fluxes at the left and right walls for cases 2, 3, and 4 are shown in Figure 5.12. The heat fluxes for cases 2, 3, and 4 follow the same pattern as case 1: the heat flux decreases quickly at the left wall, and increases gradually at the right wall, as the temperature of bulk fluid increases gradually. In Figure 5.10, some oscillations of the heat fluxes still exist for case 2, however they can hardly be observed for cases 3 and 4. This is because the thermally induced acoustic waves are too weak for cases 3 and 4, due to the slow left wall heating rates.
5.3.2 Long time solution

Since it is not computationally feasible to obtain long time solutions (order of several minutes to hours) with the explicit formulation (Courant number limitation), an implicit procedure is employed to simulate the heat transfer and flow in the enclosure filled with supercritical carbon dioxide for long time periods leading to thermal equilibrium. In this regard we employed the CFD-ACE software [92]. A time-marching implicit finite volume
method [93] is used in CFD-ACE to obtain the solutions to the governing equations with the specified boundary conditions. Euler 1st order differencing for time accuracy is chosen. For the convective-diffusive terms a second order upwind scheme [94] is used. The original idea of this second order upwind scheme is based on the evaluation of the cell face value by using linear interpolation of values at two upstream cells. Only one case is simulated with the long time solution, which is listed in Table 5.2.

Table 5.2 Case description for long time solution

<table>
<thead>
<tr>
<th></th>
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</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>310</td>
<td>312</td>
<td>310</td>
<td>7.5</td>
<td>1g</td>
<td>$2.089 \times 10^8$</td>
<td>60×60</td>
</tr>
</tbody>
</table>

Figure 5.13 shows the instantaneous velocity field for case 4 at a relative large time, $t = 100$ s. Like in the short time simulations, a recirculating flow pattern is developed in the enclosure. But the center of the recirculating flow is roughly at $(x = 0.5L, y = 0.5H)$ and the recirculating flow is symmetric about the center. Figure 5.10 also shows that although the velocity boundary layers are much thinner than those in the short time simulations, the maximum velocities are almost the same for both short time and long time simulations.
Temporal variations of pressure and temperature at the center for case 4 are shown in Figure 5.14. From it we see that equilibrium is reached at about $t = 30$ s. The equilibrium temperature at the center of the enclosure is 311 K, right at the intermediate of the left wall temperature and right wall temperature. The equilibrium temperature at the center point is a little bit above 8 MPa.
Heat transfer and flow in an enclosure filled with supercritical carbon dioxide were studied by numerically solving a fully compressible form of the Navier-Stokes equations through an explicit FCT scheme for early time and an implicit scheme for long time solutions. A highly accurate FCT algorithm, along with the theory of characteristic-based wall boundary conditions for density, was employed to simulate the early time solutions. The NIST Standard Reference Database 12 [80] was used to generate the polynomial fits to represent the equation of state and other thermodynamic properties for supercritical carbon dioxide. For the implicit scheme, CFD-ACE was employed to overcome the stability limit on $\Delta t$ to generate long time solution.

Rapid heating of the left wall generates thermally induced acoustic waves in the enclosure, which play an important role in heat transfer in supercritical fluids. Due to the
large Grashof number, recirculating flow fields developed under gravity for both initial temperature fields, which enhanced heat transfer in supercritical carbon dioxide.
CHAPTER 6

EXPERIMENTAL INVESTIGATION OF THERMALLY INDUCED ACOUSTIC WAVES

6.1 Introduction

The behaviors of thermally induced acoustic waves, in high pressure and supercritical carbon dioxide, generated by rapid heating of a bounding solid wall in a closed cylindrical tube are investigated experimentally. In the experiment, a thin aluminum foil attached to one end of the cylinder is heated by a resistance-capacitance (R-C) circuit, which is controlled by a silicon-controlled rectifier (SCR). The rapid heating of the aluminum foil generates a thermally induced acoustic wave in the closed cylinder, in which the thermally induced acoustic wave keeps traversing between the two ends and undergoes many reflections before it is fully damped out. The time-dependent pressure variations in the cylinder is measured by a microphone located at a specific point on the side of the cylinder, and the temporal temperature variations of the aluminum foil are measured by a thermocouple attached to it, along with a fast-response data acquisition system (DAQ).

6.2 Experimental apparatus

In order to generate thermally induced acoustic waves in a closed cylinder, we need to rapidly increase a wall temperature. This is a critical point for the success of current experiments, because the temperature increasing rate not only influences the character of thermally induced acoustic waves, but it also determines the strength of the waves, which determines whether the waves can be detected by a sensor.
Carbon dioxide is chosen as the working fluid, due to its good characteristics, such as non-toxicity, non-flammability, inexpensiveness, and benign critical temperature (304.13 K) and critical pressure (7.3773 MPa) [80]. After choosing Carbon dioxide as the working fluid, how to achieving supercritical state (T>304.13 K and p>7.3773 MPa) safely within the cylinder becomes crucial for the success of our experiments.

The thermally induced acoustic waves are generated and travels within a cylinder filled with supercritical carbon dioxide, while the DAQ system, the sensor cables and wires must be connected to a data logging system (a personal computer) in air. Thus how to house the pressure sensor within the supercritical carbon dioxide and how to feed the cables and wires through the supercritical carbon dioxide to the data logging system is also crucial task for our experiments.

Above are the three major concerns for our experiments. Details of the experimental apparatus will be discussed in the three following subsections:

6.2.1 Generating Thermally Induced Acoustic Waves

Based on the past experimental work [1,42], an R-C circuit is employed to rapidly heat a metal foil attached to a end of the cylinder by means of direct-current heating. The schematic of foil heating is shown in Figure 6.1. The capacitor is charged to the destination voltage at the beginning of an experiment and then it is disconnected with the charging battery. During the experiment, the triggering circuit is switched on by the relay controlled
by a triggering signal from the DAQ card. Subsequently, the capacitor is fully discharged by the silicon-control-rectifier (SCR) controlled by the triggering circuit, resulting in a rapid temperature rise in the foil due to the current flowing through it. With the same capacitance and charging voltage of the capacitor, the same amount of energy is applied to the foil, generating the same temperature rise in the foil, and, consequently, thermally induced acoustic waves with the same strength. This R-C heating system provides not only a rapid heating of the foil temperature for generating the thermally induced acoustic waves, and also some other advantages including: good repeatability and easy control of heating rate and maximum temperature increase.

![Figure 6.1 Schematic of foil heating](image)

Lin and Farouk [42] used 6.0 µm thick Nickel foils to generate thermally induced acoustic waves, because with a thinner foil, the heating is faster and the maximum temperature of the foil is higher, resulting in stronger thermally induced acoustic waves. In this way, a better measuring accuracy can be achieved. While in supercritical fluids,
thermally induced acoustic waves are much stronger than in ideal gases for the same temperature boundary conditions. This phenomena have been proved by numerical results [2,42,43] and our later experimental results. Therefore 6.0 µm thick Nickel foils is not necessary for measuring thermally induced acoustic waves in supercritical fluids, because they are expensive and vulnerable to be broken and burned. In the current study, Reynolds aluminum foils are employed to generate thermally induced acoustic waves.

In the present experimental setup, the dimensions of the foil (attached around a circular mica end-piece) are 42 mm x 50 mm x 6.0 mm. The electrical resistance of the foil is calculated to be 0.014 ohm. By carefully calculating every component in the R-C circuit, the circuit efficiency \( \varepsilon = \frac{R_{foil}}{R_{total}} \) is found to be about 53%. Several capacitors rated at 18.0, 27.0 and 56.0 mF are used for the measurements. The circuit time constant, \( \tau_{RC} = R_{total} \cdot C \) is hence 0.0005 s, 0.00071 s, and 0.0015 s for capacitances of \( C = 18.0 \) mF, 27.0 mF, and 56.0 mF used in the circuit, respectively.

The details of foil installment are shown in Figure 6.2. One of the ends of the cylinder is completely covered by the foil, which is sandwiched between two Teflon plates. The Teflon plate between the cylinder and the foil (Teflon plate I in Figure 6.2) has a hole, at its center, with the same diameter as the cylinder. The Teflon plate at the other side of the foil (Teflon plate II in Figure 6.2) is a complete round piece, on which a similar mica plate is pasted. The mica plate ensures a rigid reflecting surface for the pressure waves. The foil is anchored by two long copper bars which are in good contact with two copper power leads. The cross-sectional dimensions of the copper bars are 5 mm x 8 mm, and the diameters of the
power leads are both 5 mm. This minimizes the voltage drop through the copper bar, and also guarantees that the electric current flows through the foil evenly.

Figure 6.2 Schematic of foil installment

Figure 6.3 Schematic of experimental system
Figure 6.3 shows the schematic of experimental system. Thermally generated acoustic waves are generated and travels within a cylinder of a 1 in diameter (The cylinder will be discussed in detail in subsection 6.2.2). During an experiment, the circuit connecting the capacitor and the foil is switched on by the SCR, which is triggered by the triggering circuit.

Table 6.1 Specifications for the experimental system

<table>
<thead>
<tr>
<th>No.</th>
<th>Part</th>
<th>Manufacturer and model</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Capacitors</td>
<td>Sprague Antex A8</td>
<td>18, 27 and 56 mF</td>
</tr>
<tr>
<td>2</td>
<td>SCR</td>
<td>Littelfuse TO-218X</td>
<td>Max trigger time: 2.5 µs</td>
</tr>
<tr>
<td>3</td>
<td>Foil</td>
<td>Reynolds Aluminum</td>
<td>Nickel, 0.006 mm thick, 99.95%, resistivity 6.84x10^-8 ohm-m</td>
</tr>
<tr>
<td>4</td>
<td>Thermocouple</td>
<td>Omega ChCO-0005</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>Pressure probes</td>
<td>B &amp; K Microphone (4193)</td>
<td>2 mV/Pa</td>
</tr>
<tr>
<td>6</td>
<td>DAQ card</td>
<td>National Instrument PCI 6052E</td>
<td>333 kHz</td>
</tr>
</tbody>
</table>

6.2.2 Safe Operation of the System

Achieving supercritical state safely is one of the key points for the success of measuring thermally induced acoustic pressure waves in supercritical carbon dioxide. The critical temperature and pressure for carbon dioxide is 304.13 K and 7.3773 MPa, respectively. In order to achieving supercritical state, we need to heat the carbon dioxide from the lab room temperature (generally 295 K) to above the critical temperature, and also
to pressurize the carbon dioxide from 900 psig (the internal pressure of the CO₂ tank) to above the critical pressure.

Figure 6.4 Schematic of experimental tubing system

Figure 6.4 shows the schematic of the experimental tubing system. Carbon dioxide is stored in a tank, and in order to increase the carbon dioxide pressure, a hand pump is employed to pressurize the carbon dioxide in the supercritical chamber until the destination pressure is reached (the destination pressure is above the supercritical pressure). The reason to choose a hand pump is that it is able to compress a fluid within a small volume to develop high pressure. A hand pump also enables us to accurately regulate the pressure inside the supercritical chamber manually. Another reason is that the hand pump is easily mounted to a work bench and maximum pressures may be obtained with a minimum amount of effort. Figure 6.5 shows the schematic of the hand pump. The carbon dioxide tank (T) is
connected by means of valves and fittings to the supercritical chamber (C) that is to be pressurized.

Figure 6.5 Schematic of the hand pump

With valve “B” closed and valve “A” open, the handle of the hand pump is rotated counter-clockwise to draw carbon dioxide from the tank into the cylinder body of the hand pump. Valve “A” is then closed and valve “B” is opened. By rotating the hand pump handle clockwise, the piston will compress the carbon dioxide to develop pressure in the supercritical chamber. If sufficient pressure is not reached in one stroke, the system will be repeated. Valve “B” will be closed in order to maintain pressure in the components. Valve “A” is then opened, and carbon dioxide is again drawn into the hand pump from the tank. Closing Valve “A” and opening Valve “B” will allow the hand pump to be operated to develop increased pressure in the supercritical chamber. Figure 6.6 shows the photo of the hand pump. The Specifications of the hand pump are listed in Table 6.2.
Table 6.2 Specifications for heating and pressurizing parts

<table>
<thead>
<tr>
<th>No.</th>
<th>Part</th>
<th>Manufacturer and model</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Hand pump</td>
<td>HIP 87-6-5-Teflon</td>
<td>0-5000 psi, 60mL/strok 7/8” shaft diameter</td>
</tr>
<tr>
<td>2</td>
<td>Heating tape</td>
<td>Thermolyne .5×4'</td>
<td>533 K maximum temperature</td>
</tr>
</tbody>
</table>

The Thermolyne .5×4' heating tape is used to heat the supercritical chamber from the lab room temperature (generally 295 K) to the destination temperature (above the critical temperature). The heating tape is wrapped around the supercritical chamber and connected to its controller. Around the supercritical chamber, an adhesive tape is employed above the heating tape to make sure the entire surface of the heating tape in firm contact with the surface being heated (the outer surface of the supercritical chamber). During an experiment, when the destination temperature is reached, a lower heating power is set in the heating tape.
controller to maintain the supercritical chamber at the destination temperature. The Specifications of the heating tape are listed in Table 6.2.

With the hand pump and the heating tape, the carbon dioxide can be heated and pressurized to supercritical region. While supercritical state is reached, doing that safely and preventing explosion of the entire experimental setup during an experiment are also crucial for the success of our experiment. In order to make ensure the safety, following steps have been taken:

The supercritical chamber is well designed that it can withstand much higher pressure than the critical pressure of carbon dioxide (7.3773 MPa). Figure 6.7 shows the cross-section of the supercritical chamber. The material of the supercritical chamber is 316 stainless steel. The thickness of the thinnest wall of the supercritical chamber is 4.8 cm, which enable the supercritical chamber to withstand much higher pressure than the critical pressure of carbon dioxide. The tubes and valves are also chosen in such a way that they can withstand much higher pressure than the critical pressure of carbon dioxide. The Specifications of the tubes and the valves are listed in Table 6.3.
During an experiment, accidental pressure rise within the supercritical chamber, the tube and the hand pump should be prevented. Rupture discs are employed to make sure that the pressure within the system will never be above a specific value. By this means, once the pressure within the system is above the burst pressure the rupture discs, they will be burst and then release the high pressure carbon dioxide safely. The burst pressure the rupture discs are chosen to be 2000 psig (16 MPa), which is still a safe pressure for the experimental setup and also far away from the critical pressure of the carbon dioxide, leaving enough space for the experiments. The Specifications of the rupture discs are listed in Table 6.3.

Two rupture discs have been employed, one connected with the supercritical chamber, the other between the valves A and B (see Figure 6.4). The rupture disc connected with the supercritical chamber is used to make sure that the pressure within the chamber will not
exceed the rupture pressure of the disc. The rupture disc between the valves A and B is used to avoid extremely high pressure between these two valves. During an experiment, when the hand pump is being used to pressurize the supercritical chamber, the valves A and B will be closed alternately. When these two valves are closed simultaneously by mistake, using the hand pump will compress the carbon dioxide to develop pressure in tube between the two valves, not in the supercritical chamber. Under this circumstance, only in one stroke of the hand pump, the pressure in the tube between the two valves will reach a high pressure and make the tube burst. The rupture disc placed between valves A and B will avoid this accident.

In order to avoid blocks in the tubes, the valves, the hand pump and the sensor, a filter is placed right after the carbon dioxide tank. By this means, the carbon dioxide in the experimental setup can be made sure to be pure and clean. The Specifications of the filter are listed in Table 6.3.

<table>
<thead>
<tr>
<th>No.</th>
<th>Part</th>
<th>Manufacturer and model</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Tubing</td>
<td>HIP 87-6-5-Teflon</td>
<td>0-5000 psi, 60mL/strok</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>7/8&quot; shaft diameter</td>
</tr>
<tr>
<td>2</td>
<td>Valve</td>
<td>Thermolyne .5×4'</td>
<td>533 K maximum temperature</td>
</tr>
<tr>
<td>3</td>
<td>Rupture disc</td>
<td>ZOOK PB-RD2000 (1/4”)</td>
<td>2000 psi</td>
</tr>
</tbody>
</table>
Although many techniques are employed for the safety of the experiment system, aploy case is still built for any unexpected accident. The supercritical chamber and other probes and accessories (such as the pressure gauge, the thermocouple and rupture disc) are all placed within the case (see Figure 6.4). The case has a front slide door, which enable us to open and close the case conveniently. Out of the case are some valves, tubes, tees and the hand pump, which can withstand much higher pressure (20.7 MPa) than the critical pressure (7.38 MPa) of carbon dioxide. Also a rupture disc is installed between the two vales A and B (see Figure 6.4) to prevent any unexpected accident out of the case. Figure 6.8 shows the photo of the case with the supercritical chamber in it.

6.2.3 Mounting Pressure Sensors

Two different types of pressure sensors have been used by Lin and Farouk [42] to measure and monitor the thermally induced acoustic waves in air: a condenser microphone
(12.7 mm Brüel & Kjær model 4193) and a piezoresistive pressure transducer (Endevco model 8507C-1). The Brüel & Kjær condenser microphone was chosen because of its high sensitivity, fast response, and wide frequency-response range. The sensitivity of the Endevco pressure transducer is, however, not sufficiently good for the measurement of the small dynamic pressure fluctuations in the supercritical chamber due to the sudden heating of the foil. In figure 6.9, which shows comparison of pressure \((p – p_i)\) measurement by the Brüel & Kjær microphone (unfiltered) and Endevco pressure transducer (filtered) by Lin and Farouk [42], it can be seen that the B&K microphone yields better measurements than the Endevco pressure transducer. Thus for the experiments of thermally induced acoustic waves in supercritical carbon dioxide, the Endevco pressure transducer was discarded.

Figure 6.9 Comparison of pressure \((p – p_i)\) measurement by the Brüel & Kjær microphone (unfiltered) and Endevco pressure transducer (filtered) by Lin and Farouk [42]
For the experiments of thermally induced acoustic waves in supercritical carbon dioxide, the probes are in a high pressure zone, while the data needs to be transmitted to the DAQ card, which is in an atmospheric region. Thus how to house the probes, especially the microphone, and how to feed the probe data cable through the supercritical chamber to the DAQ card are also challenging and crucial for the success of the experiments. In the experimental setup, there are four types of parts which need be feed through the supercritical chamber: tubes, thermocouples, power leads for the foil and data cables. For these four types of parts, glands are employed to feed them through a high pressure region to a low pressure region. For different types of parts, different types of glands are used. All the specifications of the glands are listed in Table 6.4

<table>
<thead>
<tr>
<th>No.</th>
<th>Part</th>
<th>Manufacturer and model</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Tube gland</td>
<td>HIP</td>
<td>0-5000 psi, 60mL/strok</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>7/8&quot; shaft diameter</td>
</tr>
<tr>
<td>2</td>
<td>Thermocouple gland</td>
<td>HIP</td>
<td>533 K maximum temperature</td>
</tr>
<tr>
<td>3</td>
<td>Power lead gland</td>
<td>HIP</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>Cable gland</td>
<td>HIP</td>
<td></td>
</tr>
</tbody>
</table>

Figure 6.10 Photo of B&K 4193 microphone (left) and its preamplifier (right)
Housing the pressure probe, B&K 4193 microphone, is the most critical issue for the success of the experiment. Figure 6.10 shows the photo of B&K 4193 microphone and its preamplifier. In order to measure the thermally induced acoustic pressure waves in the supercritical chamber, the diaphragm (right under the grille in Figure 6.10) of the microphone is placed flush with the inner surface of the supercritical chamber. The microphone is connected with the preamplifier through a screw thread on the preamplifier. A cable connects the preamplifier and the amplifier, which is connected with the DAQ card. So during an experiment, the microphone is in the supercritical region, while the cable connects the microphone and preamplifier, which are in the supercritical region, with the amplifier and the DAQ card, which are in room air. Thus how to house the microphone with the diaphragm flush with the inner surface of the supercritical chamber and how to feed the cable from the supercritical region to room air are the two main problems for successfully housing the pressure probe.

![Figure 6.11 Schematic of microphone housing](image_url)
6.3 Experimental Procedure

At the beginning of an experiment, after powering on and warming up, all setups and devices are checked for the reliability of the experiment. The capacitor is charged by a power source to a desired voltage $V_0$, which is measured by an HP 34401A multi-meter. The SCR is triggered to initiate the experiment, and the capacitor consequently is discharged, causing the rapid heating of the foil. The foil temperature gradually falls after an initial rapid rise. The Labview 8.0 software is used to record the foil temperature and pressure in the experiments, and to provide controlling signals. For most experiments, data are collected for about 4 seconds after the capacitor is discharged. After one experiment is carried out, the experimental conditions are initialized before the next measurement. Each experiment is repeated several times in order to confirm repeatability of the measurements.

6.4 Experimental Results and Discussion

6.4.1 Thermally Induced Acoustic Waves in Supercritical (High Pressure/High Temperature) Nitrogen

Some experiments were first carried out in Nitrogen at initial conditions: $p_i = 3.55$ MPa and $T_i = 297$ K ($24^\circ$C). The critical pressure and temperature of Nitrogen is: $P_c = 3.4$ MPa and $T_c = 126.2$ K, respectively. Nitrogen was chosen first due to its low critical pressure and thus the less possibility destroying the microphone. A 56 mF capacitor was used during the measurements. For this given capacitor, a number of different charging voltages $V_0$ were applied.
Figure 6.12 Temporal variation of the foil temperature ($V_0 = 35$ V)

The foil temperature ($V_0 = 35$ V) measured by the thermocouple is shown in Figures 6.12 (a) and 6.12 (b). At early times (for less than 0.02 s after the beginning of capacitor discharging) the foil temperature increases rapidly, but the increasing rate decreases gradually. The temperature rise in the foil depends on the heat gain and loss to the supercritical nitrogen. After the capacitor discharge period, the foil temperature began to decrease gradually due to the heat loss to the supercritical nitrogen. It is also noted that during the capacitor discharge period, the thermocouple cannot accurately measure the foil temperature during the initial discharge period because of the severe electromagnetic disturbance caused.
Figure 6.13 Temporal variation of pressure \((p - p_i)\) wave for \((V_0 = 35 \text{ V})\)

The temporal variation of the pressure wave \((V_0 = 30 \text{ V})\) measured by the Brüel & Kjær microphone is shown in Figures 6.13 (a) and 6.13 (b). When the pressure wave arrives at the location where the microphone is installed, the pressure at that location undergoes rapid temporal variations, which is measured and recorded by the probe. As the pressure wave is travelling between the side walls of the cylinder, hundreds of pressure peaks are measured by the probe before the wave has been damped out. During the first several acoustic cycles (Figure 6.13 (a)) the wave shape shows steep fronts followed by some noises, which is caused by the uneven foil surface and unsmooth internal surface of the cylinder. Due to viscous dissipation and energy losses, the sharp wave peak gets flatter and flatter, and the wave amplitude also decreases gradually. The measured acoustic speed well matches the value predicted by the NIST Standard Reference Database 12 [80].
The pressure variation recorded by the Brüel & Kjær 4193 microphone over a longer period of time is presented in Figure 6.13 (b). It is interesting to observe that the probe records pressure values lower than the ambient value (which is non-physical values) beginning at about $t = 0.4$ s. The anomalous result is caused by the inherent construction and operational features of the Brüel & Kjær condenser microphone. The existence of the narrow vent hole (see Figure 6.14) causes the non-physical results at longer times. Thus the Brüel & Kjær microphone is not suitable for the measurement with varying static pressure, while it is good for the measurement of dynamic pressure where static pressure remains constant. When the foil is first heated, it heats the gas inside the tube, so the static pressure increases. Therefore, the static pressure inside the tube increases to a maximum value, then decreases to the initial value in several seconds as the heat is lost to the outside environment from the wall. As the diameter of the vent hole connecting the microphone housing to the pressure field in the tube is very small, the static pressure inside the microphone housing varies more slowly than that in the enclosed pressure field. Within some time period, the static pressure
inside the microphone housing becomes greater than that of the air in the tube, causing the pressure readings to fall below zero (from \( t = 0.4 \) s to \( t = 2 \) s in Figure 6.13 (b)). Finally, the static pressure difference between the microphone housing and the tube is eliminated (due to the cooling of the system), and the pressure measurement will become equal to zero, as displayed in Figure 6.13 (b).

A series of measurements were carried out based on different capacitor charging voltages. The effect of capacitor charging voltage on the foil temperature is shown in Figure 6.15. As the charge voltage of the capacitor only affect the amount of electric energy stored in the capacitor, the characteristics of the foil temperature remain the same for different charging voltages.

![Figure 6.15 Temporal variations of the foil temperature (showing the effect of the capacitor charging voltage)](image)
Figure 6.16 Temporal variations of pressure ($p - p_i$) wave (showing the effect of the capacitor charging voltage)

Figure 6.16 shows the effects of the capacitor charging voltage on the acoustic wave. Stronger acoustic waves are generated for higher charge voltages, because of the higher temperature increases rate. Also, the static pressure increase is higher for higher charge voltage. Physically, the thermally induced acoustic wave is generated by the local pressure disturbance near the wall due to the sudden gas temperature increase. The sudden gas temperature increase is caused by the electric current in the foil when the capacitor is being discharged. Therefore, the strength of the thermally induced acoustic wave is found to vary with the capacitor charging voltage.

6.4.2 Thermally Induced Acoustic Waves in Supercritical Carbon Dioxide

Experiments were first conducted in supercritical nitrogen ($P_c = 3.4$ MPa and $T_c = 126.2$ K), due to the low critical pressure. While at the experimental initial conditions ($p_i =$
3.55 MPa and Ti = 297 K), the property behaviors are like those of ideal gases, due to the much higher experimental initial temperature compared with the critical temperature of nitrogen. Therefore, experiments were then carried out in supercritical carbon dioxide (P_c = 7.38 MPa and T_c = 304.13 K). Although the high critical pressure of carbon dioxide increase the risk destroying the microphone, the critical temperature, which is quite close to the room temperature, makes the experimental initial conditions (p_i = 7.69 MPa and Ti = 307 K) nearer to the critical point in the supercritical region. Under this initial state, the thermophysical properties exhibit the typical behaviors for supercritical fluids. At the beginning of an experiment, the supercritical chamber was slowly heated by a heating tape, which is wound round the chamber, to the initial temperature (Ti = 307 K). After the desired temperature was reached, the power of the heating tape was adjusted in order to maintain the desired initial temperature. Before the capacitor was discharged to generate thermally induced acoustic waves, the supercritical chamber was maintained at the initial temperature for some time to ensure steady state within it.

Figure 6.17 shows the temporal variation of the foil temperature (V0 = 35 V). Similarly as the foil temperature in supercritical nitrogen, the foil temperature increases rapidly to the maximum value at early times (for less than 0.04 s after the beginning of capacitor discharging). However, the maximum temperature rise of the foil in the supercritical nitrogen (p_i = 3.55 MPa and Ti = 297 K) is much stronger than that in the supercritical carbon dioxide (p_i = 7.69 MPa and Ti = 307 K) for the same capacitor charging voltage. This can be explained by the higher volumetric heat capacity (the product of ρc_p) for the supercritical carbon dioxide. Also, it is also noted that during the capacitor discharge period, the severe
electromagnetic disturbance caused noises and inaccurate data in the thermocouple measurements.

(a) Early time                                            (b) Longer time scale

Figure 6.17 Temporal variation of the foil temperature ($V_0 = 35$ V)

(a) Early time                                            (b) Longer time scale

Figure 6.18 Temporal variation of pressure ($p - p_i$) wave ($V_0 = 35$ V)
The Temporal variation of pressure \((p - p_i)\) wave \((V_0 = 35 \text{ V})\) in the supercritical carbon dioxide is shown in Figure 6.18. During the first several acoustic cycles (Figure 6.18 (a)) the wave shape shows the same steep fronts followed by some noises as in the supercritical nitrogen. The noises are still caused by the uneven foil surface and unsmooth internal surface of the cylinder. It is interesting to note that the same capacitor charging voltage generates much stronger thermally induced acoustic waves in the supercritical carbon dioxide than in supercritical nitrogen, due to the higher \(\left(\frac{\partial p}{\partial T}\right)_v\) for the supercritical carbon dioxide. Additionally, the measured acoustic speed is in a good match with value predicted by the NIST Standard Reference Database 12 [80]. In Figure 6.18, another interesting phenomenon was observed that the pressure profile exhibit low frequency disturbances in both early time or longer time figures. This phenomenon is caused by the fluid flow within the supercritical chamber induced by the non-uniform temperature field caused by the heating tape.

![Graph showing temporal variations of pressure](image)

(a) Early time  
(b) Longer time scale

Figure 6.19 Temporal variations of pressure \((p - p_i)\) wave (showing the effect of the capacitor charging voltage)
The effect of capacitor charging voltage on the foil temperature was also studied in the supercritical carbon dioxide experiments (\(p_i = 7.69\) MPa and \(T_i = 307\) K). Figure 6.19 shows the temporal variations of pressure (\(p - p_i\)) wave for different capacitor charging voltages. In Figure 6.19 (a), we see that the wave speed and the wave shape are the same for different capacitor charging voltage. At the early time the wave amplitude is found to be dependent on the foil temperature increasing rate, which is determined by the capacitor charging voltage.

**6.5 Estimation of Experimental Uncertainty**

Several independent measurements were conducted in this section. The measurements in the experiments include: pressure in the supercritical chamber measured by the Brüel & Kjær 4193 microphone, foil temperature measured by the a thermocouple. All these probes are connected with the NI 6052E DAQ card. The accuracy of the NI 6052E is \(\pm 0.242\) mV for the range of voltage measurements made by the 4193 microphone, and \(\pm 0.045\) ºC for the thermocouple measurements.

The uncertainty for Brüel & Kjær 4193 microphone is \(\pm 0.2\) dB and for the 2690 signal conditioner of the microphone it is \(\pm 0.1\) dB. The total maximum error for the pressure measurements made by the Brüel & Kjær 4193 microphone is estimated to be \(\pm 0.15\) Pa, including the error introduced by the DAQ board. In the thermocouple measurements, the cold junction compensation has an error of \(\pm 0.045\) ºC, error from the amplifier is \(0.01\%\) ºC and the converting equation also introduces an error of \(\pm 0.02\) ºC error. Thus the total temperature error is \(0.145\) ºC plus \(0.01\%\) of the reading value, including the \(\pm 0.08\) ºC error from DAQ card.
The transient pressure measurements by the Brüel & Kjær 4193 microphone exhibit very high degrees of consistency and very low uncertainties. For the 4193 microphone measurement, the uncertainty, with a 95% confidence level, is 0.75% for the experimental case with the lowest charging voltage and capacitance. The maximum uncertainty is ±1.0% for the temperature measurements.

6.6 Comparison of Numerical and Experimental Results

Comparison of numerical and experimental results at early time was carried out in this section. Comparison of numerical and experimental results at longer time was not performed, because the flow field caused by the heating tape and foil strongly affects the pressure wave measurements and makes the comparison difficulty and meaningless.

Figure 6.20 Comparison of experimental and computational temporal variations of pressure at the center between the left and right walls in supercritical carbon dioxide (V₀ = 35 V)
In order to save computational time, a 0.01-meter-long 1d problem geometry was employed for the numerical simulation, although the length of the cylinder filled with supercritical fluids in experimental is 0.257 m. A 1d problem geometry was employed because the problem is essentially one-dimensional at early time. The mesh size is 500 and the Courant number is 0.4 for the numerical simulation. In numerical simulations, setting the temperature boundary condition for the heated wall is rather challenging, because the characteristics of thermally induced acoustic waves are determined by the initial heating rate at the right beginning of the heating process. However at the right beginning of the heating process, the temperature measurement by the thermocouple was strongly affected by the severe electromagnetic disturbances generated by the severe temporal variation of electric current in the foil. In the numerical model, the temperature boundary condition is determined by an exponential heating equation with the same initial heating rate as in the experimental measurements.

Figure 6.20 compares the experimental and computational pressure waves \((V_0 = 35 \text{ V})\). In Figure 6.20 the vertical axis is pressure \((p - p_i)\) and the horizontal axis is the non-dimensional time \(t^*c/L\), where \(t^* = t/c\), \(c\) is acoustic speed and \(L\) is length. The pressure wave speed and amplitude are correctly predicted in the numerical results, the same as the experimental results. Furthermore, as the wave is travelling between the left and right walls, the pressure rise of bulk fluid is well predicted by the numerical model. However, as there is much noise after each wave peak in the temporal pressure variation in the experimental results, the pressure rise of bulk fluid is not as clear as that in the numerical results at early time. But the pressure rise of bulk fluid is still quite evident in experimental results at long
time scale (see Figure 6.19). Another difference between the experimental and numerical results is: each wave peak in the experimental results is much wider than in the numerical results. This can be explained by the large diaphragm diameter (roughly 1 cm) of the B&K microphone (Type 4193) compared with the small cell length ($2\times10^{-5}$ m) of the numerical model. The B&K microphone actually measures the average pressure within the region in contact with its diaphragm, while the numerical results show the pressure right at the center cell of the computational domain.

6.7 Conclusions

Experimental measurements were conducted to characterize the generation, propagation and damping of thermally induced acoustic waves in supercritical nitrogen ($p_i = 3.55$ MPa and $T_i = 297$ K) and supercritical carbon dioxide ($p_i = 7.69$ MPa and $T_i = 307$ K). In the experiments, the generation, propagation and damping of pressure waves for different capacitor charging voltage were investigated, and the strength of the thermally induced acoustic wave was found to be dependent on the temperature increase rate of the foil at early time. The maximum foil temperature rise in the supercritical nitrogen is also found to be higher than in the supercritical carbon dioxide, due to the lower volumetric specific heat ($\rho c_p$) in the supercritical nitrogen. Low frequency disturbances was observed in both early time and longer time pressure profiles, due to the fluid flow within the supercritical chamber induced by the non-uniform temperature field caused by the heating tape. At early times, the computational and experimental results are in a qualitative match.
7.1 Introduction

Interaction of acoustic waves in compressible fluids and solid boundaries creates challenging problems. It is well-known that sound sources may generate flow fields in which the particle velocities are not simply sinusoidal, and a pattern of time-independent vertical flows or steady circulations is often observed in the body of compressible media [95,96]. These second-order steady flow patterns are known as “acoustic streaming”. The steady flow velocities increase as the intensity of the sound sources increases, however the secondary streaming velocity magnitude is much lower than the primary oscillatory particle velocity magnitude, even at high sound source intensity levels. Acoustic streaming can be classified on the basis of the different mechanisms by which the streaming is generated [97]:

Rayleigh streaming inside a standing wave resonator is referred to as “boundary layer driven” streaming, as the induced flow is caused by the shear viscosity close to a solid boundary [98]. Rayleigh streaming is often called “outer streaming”, because it describes the mean fluid flow outside the boundary layer. Schlichting streaming, the mean flow motion inside the boundary layer of a standing wave field, is also known as “inner streaming. Inner and outer streaming can be formed inside a channel where the acoustic wave propagates longitudinally (Figure 7.1) or there is oscillation of solid boundary wall inside the bulk medium (Figure 7.2).
Figure 7.1 Schematic of acoustic streaming in a channel [97]

Figure 7.2 Schematic of acoustic streaming induced by oscillation of cylinder [99]
Eckart streaming is driven by dissipation within the fluid volume and is predominant in the high frequency range [98]. This kind of streaming has been used in ultrasonic applications and can be referred to as “bulk dissipation driven” streaming.

“Jet-driven streaming” is another streaming mechanism. It is associated with the periodic suction and ejection of a viscous fluid through an orifice or a change in resonator cross section [98]. The mechanism of jet driven streaming relies on the fact that a viscous fluid behaves quite differently during the suction and ejection periods. During suction, the flow in the orifice comes effectively from all directions. However, in the ejection period, a jet is generated that induces a mean flow in a certain direction. The vortices in the jet shear layer in the vicinity of the orifice or change in cross sectional area are responsible for this kind of streaming. Finally, “traveling wave streaming” occurs in the presence of a progressive wave and exhibits certain unique features.

Acoustic streaming may be effective in accelerating certain kinds of rate processes, especially the cooling process of electronic systems under micro-gravity conditions, where nature convections are greatly reduced or completely eliminated. Such secondary streaming flows can also be employed to enhance mixing processes and to augment heat and mass transfer through resonator walls. Acoustic streaming can be an effective technique for enhancing convection within micro- and zero-gravity environments, where conduction is the leading or only mode of heat transfer [100].
While various analytical models, numerical models and experimental results are available for studying acoustic streaming phenomenon, all of them are limited in ideal gases or liquids. For analytical and numerical models in ideal gases, they are usually based on the fact that internal energy is a function of temperature only, which is not valid any more for supercritical fluids. In this chapter, acoustic streaming motion generated by finite-amplitude resonant oscillations in a two-dimensional enclosure, filled with supercritical nitrogen, is numerically simulated. Other than the formation of acoustic streaming in supercritical nitrogen, the pressure and temperature variations in different initial states are also investigated numerically.

### 7.2 Studies of Mechanically Driven Acoustic Streaming in ideal gases

The study of acoustic streaming started with the theoretical work of Lord Rayleigh [101]. He considered vortex flows occurring in a long pipe (Kundt’s tube) as a result of the presence of a longitudinal standing wave. Westervelt [102] obtained a general vorticity equation and developed a general procedure for evaluating the streaming velocity induced by acoustical disturbances. After reviewing the theories for calculating steady streaming associated with sound fields, Nyborg [103] studied two illustrative problems, both for rectilinear flow due to irrotational sound fields: the first deals with a single attenuated plane wave traveling down a tube, and the second deals with a pair of crossed plane waves. Nyborg also developed an approximate solution for sonically-induced steady flow near a fluid-solid interface subject to a condition of a known irrotational oscillatory velocity distribution in the vicinity of the interface.
The effect of compressibility on acoustic streaming near a rigid boundary was theoretically studied by Qi [20]. Qi extended previous analyses of acoustic streaming, which were limited to incompressible fluids, to compressible fluids. Qi found that the inclusion of compressibility leads to a larger streaming velocity outside the boundary layer in gases, but not in liquids. Qi [104] also theoretically investigated the effect of compressibility and heat conduction on the viscous and thermal boundary layer attenuation and the acoustic streaming that accompanies the lowest acoustic mode propagating down a circular tube. However Hamilton et al. noted that the physical interpretation of acoustic streaming proposed by Qi was misleading, because Qi’s solution reduced to Rayleigh’s solution in the absence of heat conduction. Hamilton also revealed that that for channel widths 10–20 times the viscous penetration depth, thermal effects may alter the streaming velocity substantially. For significantly wider or narrower channels, thermal effects influence the streaming velocity by only a few percent.

Vainshtein [105] combined the problems of Rayleigh streaming and Couette flow, and investigated the effect of streaming on shear flows. Menguy and Gilbert [106,107] investigated the Nonlinear acoustic streaming in cylindrical guides. A noticeable distortion of the acoustic field due to the fluid inertia was revealed, a comparison of slow and nonlinear acoustic streaming was presented. Hamilton et al. [18] derived an analytic solution for acoustic streaming generated by a standing wave in a viscous fluid that occupies a two-dimensional channel of arbitrary width. In their study, both the outer, Rayleigh streaming vortices and the inner, boundary layer vortices are accurately described. They revealed that as channel width is reduced, the inner vortices increase in size relative to the Rayleigh
vortices. For channel widths less than about 10 times the boundary layer thickness, the Rayleigh vortices disappear and only the inner vortices exist.

Richardson [99] analytically investigated the effect of acoustic streaming on convection from a circular cylinder subjected to transverse oscillations relative to fluid in which it is immersed. Kawahashi and Arakawa [108] carried out a numerical analysis of acoustic streaming induced by finite-amplitude oscillation of air column in closed duct, using the fourth-order spatial difference method. Kawahashi and Arakawa obtained results showing velocity distributions in the oscillatory boundary layer and structures of steady streaming for various amplitudes of oscillation. Gopinath and Mills [109] analytically and numerically investigated convective heat transfer due to acoustic streaming across the ends of a tube supporting a standing sound wave. In this study, a parametric study of the effects of the controlling acoustic and geometric variables was conducted, and Nusselt number correlations were developed for air.

Yano [110] numerically investigated acoustic streaming induced by resonant gas oscillations with periodic shock waves in a closed tube by a finite-difference method. The numerical simulations revealed that relatively strong vortices are localized near the tube wall, which are quite different from the results of Rayleigh streaming, for low streaming Reynolds numbers ($= \pi X_{\text{max}} c_0 / \nu$). For medium streaming Reynolds numbers acoustic streaming involves unsteady vortices in a region near the center of the tube. For high streaming Reynolds numbers, turbulent acoustic streaming appear in the form of irregular vertical flow structures throughout the tube. Alexeev and Gutfinger [111] experimentally and numerically
studied periodic gas oscillations in closed tubes. They considered a two-dimensional flow in a compressible gas with oscillations accompanied by shock waves traveling back and forth along the tube. It is revealed that direction of gas streaming at resonance is opposite to that in nonresonant oscillations.

In above literature review, in most of the analytical and numerical studies cited in the literature, the formation of acoustic streaming is analyzed with a simplified approach. In these studies, a perfectly sinusoidal primary standing wave field in the domain is assumed and the investigation is based on this assumption. Aktas and Farouk [96] numerically investigated acoustic streaming motion in a compressible gas filled two-dimensional rectangular enclosure. Yiqiang and Farouk [112] numerically studied the effect of acoustic streaming on heat transfer in a gas-filled enclosure with differentially heated horizontal walls. In above numerical works, no pre-defined sound field in the computational domain is employed for the simulation of acoustic streaming. A rectangular enclosure filled with compressible gas is considered as their domain. The left wall, modeled as a rigid boundary, vibrates harmonically in time representing the motion of a loud-speaker diaphragm or vibration of a commercial ultrasonic mixer probe. The vibrating wall is the acoustic source that creates a sound field in the enclosure.

7.3 Studies of Acoustically Enhanced Transport in Supercritical Fluids

As discussed in section of introduction (Section 7.1), various researchers have analytically, numerically and experimentally investigated the acoustic streaming phenomenon, all works are limited in ideal gases or liquids. As supercritical fluids are more
and more widely used in various industrial and laboratory processes, acoustically enhancement of transport in supercritical fluids begins to attract researching interest. Riera et al. [72] experimentally investigated mass transfer enhancement in supercritical fluids extraction by means of power ultrasound. Riera et al. studied a supercritical carbon dioxide extraction of oil from particulate almonds. To examine the effect of the acoustic waves all experiments were performed with and without ultrasound.

In the experimental setup of Riera et al., a power ultrasonic transducer with a working frequency of 20 kHz was installed inside a high pressure supercritical fluid extractor. The experiments were carried out with carbon dioxide at 28 MPa and 55°C. Grounded almonds with an oil content of about 55% were deposited inside the supercritical fluid reactor. The power ultrasonic system developed has shown to be very efficient for the generation of ultrasonic energy under supercritical conditions. The experimental results showed that the kinetics and the extraction yield of the oil were enhanced by 30% and 20% respectively, when a power of about 50 W was applied to the transducer. Riera et al. believed that these improvements may be basically attributed to an increase in the mass transfer coefficient.

7.4 Mechanically Driven Acoustic Waves in Supercritical Fluids

7.4.1 Problem Geometry

A two-dimensional rectangular enclosure filled with supercritical nitrogen with thermally insulated and no-slip walls is considered as the computational domain, which is shown in Figure 7.3. The left wall of the enclosure vibrates harmonically, generating acoustic waves in this geometry. The harmonic displacement and the velocity of the vibrating wall are
given as \( x = X_{\text{max}}\sin(\omega t) = X_{\text{max}}\sin(2\pi ft) \) and \( v_w = \omega X_{\text{max}}\cos(\omega t) = \omega X_{\text{max}}\cos(2\pi ft) \), respectively. Here \( f \) is the vibration frequency, \( X_{\text{max}} \) is the maximum displacement, \( \omega = 2\pi f \) is the angular frequency, and \( \omega X_{\text{max}} \) is the maximum velocity of the left wall. The vibration frequency of the left wall is chosen as \( f = 20 \text{ kHz} \) or \( 10 \text{ kHz} \), as listed in Table 7.1. The corresponding wavelength of the sound waves is \( \lambda = \frac{c_0}{f} \), where \( c_0 \) is the acoustic speed. The length of the enclosure is chosen as \( L = \lambda/2 \) for all cases studied. The length of the enclosure is different for different cases with different initial stats and frequencies, as listed in Table 7.1.

The height of the enclosure is chosen as \( H = 100\delta_v \) for all cases in Table 7.1. Here \( \delta_v \) denotes the acoustic boundary layer thickness, is defined by \( \delta_v = (2v/\omega)^{0.5} \), where \( v \) is the kinematic viscosity of the fluid.

To simulate accurately the formation of acoustic streaming structures and heat transfer along the walls, resolving the boundary layer in the computational method is essential. For this reason, non-uniform grid structures were employed. This structure has finest grid in the vicinity of the walls and the grid quality gradually decreases as the distance from the horizontal walls increases. For the present calculations, there are always 3 to 5 cells inside the boundary layer. The mesh size used for the rectangular domain varies from \( 300 \times 160 \) to \( 220 \times 140 \) for different cases, as listed in Table 7.1. To model the wall vibration, moving cell boundaries were considered for a few cells next to the left wall. The number of moving cell is chosen such that the maximum variation of a cell size is less than 25%. The
governing equations (except for the diffusion terms) are discretized and solved using an explicit flux-corrected transport algorithm (LCPFCT). The timestep is determined by setting the Courant number to be 0.6.

Figure 7.3 Schematic of the computational domain

Table 7.1 Cases for parametric study of acoustic streaming in supercritical nitrogen

<table>
<thead>
<tr>
<th>Case #</th>
<th>(T_i) [K]</th>
<th>(P_i) [MPa]</th>
<th>(L\times H) [mm×mm]</th>
<th>(L/H)</th>
<th>(f) [kHz]</th>
<th>(\delta_v) [µm]</th>
<th>(H/\delta_v)</th>
<th>(X_{\text{max}}) [µm]</th>
<th>Mesh size</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>300</td>
<td>3.80</td>
<td>9.009×0.2625</td>
<td>34.3</td>
<td>20</td>
<td>2.625</td>
<td>100</td>
<td>1</td>
<td>300×160</td>
</tr>
<tr>
<td>2</td>
<td>300</td>
<td>3.80</td>
<td>9.009×0.2625</td>
<td>34.3</td>
<td>20</td>
<td>2.625</td>
<td>100</td>
<td>0.3</td>
<td>300×160</td>
</tr>
<tr>
<td>3</td>
<td>140</td>
<td>3.80</td>
<td>10.490×0.1677</td>
<td>62.6</td>
<td>10</td>
<td>1.677</td>
<td>100</td>
<td>3</td>
<td>300×160</td>
</tr>
<tr>
<td>4</td>
<td>127</td>
<td>3.52</td>
<td>7.590×0.1368</td>
<td>55.5</td>
<td>10</td>
<td>1.368</td>
<td>100</td>
<td>3</td>
<td>220×140</td>
</tr>
<tr>
<td>5</td>
<td>126.8</td>
<td>3.49</td>
<td>7.431×0.1385</td>
<td>53.7</td>
<td>10</td>
<td>1.385</td>
<td>100</td>
<td>2</td>
<td>220×140</td>
</tr>
<tr>
<td>6</td>
<td>126.8</td>
<td>3.49</td>
<td>7.431×0.277</td>
<td>26.8</td>
<td>10</td>
<td>1.385</td>
<td>200</td>
<td>1</td>
<td>220×222</td>
</tr>
</tbody>
</table>
$T_c = 126.2 \text{ K} \quad P_c = 3.4 \text{ MPa}$ for nitrogen

### 7.4.2 Results and discussion

The simulations were started with the vibration of the left wall at $x = 0$ and with uniform initial pressure and temperature fields within the quiescent supercritical fluid. Figure 7.4 shows the spatial variations of pressure along the symmetry axis of the enclosure at four different instants ($\omega t = 0, \pi/2, \pi, 3\pi/2$) during the 90th cycle for case 1. The flow is found to be quasi-steady after the 90th cycle. At $\omega t = 0$ and $\omega t = \pi$, the pressure wave in the enclosure reaches a maximum amplitude. At the beginning of the cycle ($\omega t = 0$), the pressure is maximum on the vibrating wall ($x = 0$) of the enclosure and decreases with the distance from the vibrating wall until the minimum is reached on the right wall ($x = L$). In the middle of the cycle ($\omega t = \pi$), the pressure profile is fairly symmetric to the profile at the beginning of the cycle ($\omega t = 0$), with respect to the vertical midplane ($x = L/2$). The pressure profiles at $\omega t = \pi/2$ and $\omega t = 3\pi/2$ follows the similar patterns as the profiles at $\omega t = 0$ and $\omega t = \pi$, respectively, but with smaller pressure wave amplitudes. In Figure 7.4, the pressure profiles at different times intersect approximately at $x = L/2$, thus creating a pressure node. From Figure 7.4, we see that the roughly sinusoidal and symmetric pressure wave profiles in the enclosure are slightly distorted due to viscous and nonlinear effects.

The spatial variations of $u$ velocity along the symmetry axis of the enclosure at four different instants ($\omega t = 0, \pi/2, \pi, 3\pi/2$) during the 90th acoustic cycle for Case 1 is shown in Figure 7.5. Unlike the pressure profiles, the velocity maximums and minimums appears at $\omega t = \pi/2$ and $\omega t = 3\pi/2$. The velocity of the vibrating wall is zero at $\omega t = \pi/2$ and $\omega t = 3\pi/2$ and...
reaches the maximum value at $\omega t = 0$ and $\omega t = \pi$. The primary oscillatory flow is periodic and the maximum value appears in small region around $x = L/2$. In Figure 7.5, due to the viscous and nonlinear effects, the $u$ velocity profiles are also slightly distorted as the pressure profiles in Figure 7.4.

Figure 7.4 Spatial variations of pressure along the symmetry axis of the enclosure at four different instants ($\omega t = 0, \pi/2, \pi, 3\pi/2$) during the 90$^{th}$ cycle for case 1.
Figure 7.5 Spatial variations of u velocity along the symmetry axis of the enclosure at four different instants ($\omega t = 0, \pi/2, \pi, 3\pi/2$) during the 90th acoustic cycle for Case 1.

The predicted secondary flow field for case 1 at the 90th cycle is shown in Figure 7.6. The flow field is calculated based on the time averaging method. Comparing Figure 7.5 with Figure 7.6 shows that the maximum streaming velocity is greatly smaller than the maximum instantaneous velocity in the primary oscillatory flow fields in the enclosure. The maximum streaming velocity value is found to be approximately 0.015 m/s while the maximum instantaneous velocities reach 3.5 m/s in the primary oscillatory flow field in the enclosure. In Figure 7.5, four outer streaming circulations (two clockwise and two counterclockwise) are observed in the enclosure, however no inner streaming circulation is observed in the acoustic boundary layers along the top and bottom walls. The horizontal length of all the outer streaming vortices is predicted to be quarter-wavelength ($\lambda/4$).

Figure 7.6 Mean flow field in the enclosure at the 90th acoustic cycle for case 1.
In the next case simulated (case 2), the maximum vibrating wall displacement is decreased to $X_{\text{max}} = 0.3 \, \mu\text{m}$, and the height is kept the same ($H/\delta_v = 100$). Figure 7.7 shows the spatial variations of pressure along the symmetry axis of the enclosure at four different instants ($\omega t = 0, \pi/2, \pi, 3\pi/2$) during the 90$^{\text{th}}$ cycle for case 2. Comparing Figure 7.7 with Figure 7.4 shows that the pressure waves generated by the sinusoidal displacement of the vibrating wall are less distorted with lower amplitudes in the enclosure for case 2. With a higher vibrating wall displacement value in case 1 ($X_{\text{max}} = 1 \, \mu\text{m}$), the pressure wave form is much attenuated and the pressure wave amplitude reaches approximately 100 kPa. For a smaller vibrating wall displacement value in case 2 ($X_{\text{max}} = 0.3 \, \mu\text{m}$), the pressure wave profile is near-sinusoidal and the pressure wave amplitude is 30 kPa.

![Figure 7.7 Spatial variations of pressure along the symmetry axis of the enclosure at four different instants ($\omega t = 0, \pi/2, \pi, 3\pi/2$) during the 90$^{\text{th}}$ cycle for case 2.](image-url)
The Figure 7.8 compares the temporal variations of pressure for case 1 and case 2 at the center point of the enclosure. The amplitude of pressure oscillation at the center point of the enclosure is approximately 25 kPa for 1 µm maximum wall displacement and 3 kPa for 0.3 µm maximum wall displacement. It is interesting to note that a stronger peak and a weaker peak of the pressure oscillation at the center point of the enclosure appears alternatively in Figure 7.8, due to the distorted pressure wave profile in the enclosure caused by viscous and nonlinear effects. This phenomenon is stronger for a higher wall displacement value (1 µm) than that for a lower wall displacement value (0.3 µm). The corresponding quasi-steady mean (time-averaged) flow field in the enclosure is shown in Figure 7.9. The maximum primary oscillatory flow velocity reaches approximately 1 m/s while the maximum mean flow velocity is 0.002 m/s in the secondary steady streaming flow field. In the enclosure, four outer streaming circulations (two clockwise and two counterclockwise) are clearly visible in the enclosure. Comparing the mean flow fields for case 1 (Figure 7.6) and for case 2 (7.9), we see that decreasing the maximum vibrating wall displacement does not affect the streaming structure, but the strength of the secondary steaming flow field, for the geometry of these cases. Furthermore, inner acoustic streaming is not observed in the enclosure for both cases 1 and 2.
Figure 7.8 Temporal variations of pressure at the center point of the enclosure for case 1 \((X_{\text{max}} = 1 \, \mu\text{m})\) and case 2 \((X_{\text{max}} = 0.3 \, \mu\text{m})\).

Figure 7.9 Mean flow field in the enclosure during the 90th acoustic cycle for case 2.

In the next two cases (case 3 and 4), we consider two initial states (case 3: 140 K and 3.8 MPa, case 4: 127 K and 3.52 MPa) closer to the critical point. For these two cases we
consider the same $X_{\text{max}} = 3 \, \mu\text{m}$ and $H/\delta v = 100$. The mean flow fields in the enclosure for case 3 (during the 50th acoustic cycle) and case 4 (during the 40th acoustic cycle) are presented in Figure 7.10. The secondary streaming flow fields are much weaker than the primary oscillatory flow fields in the enclosure for cases 3 and 4, respectively. The maximum velocities are approximately $0.025 \, \text{m/s}$ in the secondary streaming flow fields for both cases 3 and 4. The stronger secondary streaming flow fields in cases 3 and 4 ($X_{\text{max}} = 3 \, \mu\text{m}$) than those in case 1 ($X_{\text{max}} = 1 \, \mu\text{m}$) and case 2 ($X_{\text{max}} = 0.3 \, \mu\text{m}$) are caused by the higher maximum vibrating wall displacement in case 3 and 4. The same acoustic streaming structure without inner acoustic streaming are also observed in the enclosure for cases 3 and 4, as for cases 1 and 2.
In the next two cases (case 5 and 6), we consider the same initial state (126.8 K and 3.49 MPa), but closer to the critical point. For this initial state we consider $X_{\text{max}} = 2 \, \mu m$ (case 5) and $X_{\text{max}} = 1 \, \mu m$ (case 6) as the maximum vibrating wall displacement, $H/\delta v = 100$ (case 5) and $H/\delta v = 200$ (case 6) as the enclosure height. Figure 7.11 compares the Spatial variations of pressure along the symmetry axis of the enclosure at four different instants ($\omega t = 0, \pi/2, \pi, 3\pi/2$) during the 40th cycle for cases 5 and 6. The pressure wave generated by the vibrating
wall in the enclosure is less distorted for case 5 than for case 6. In Figure 7.11 (a), the pressure profiles at four instants for case 5 are more sinusoidal and intersect in a small region around $x = L/2$ while they are more irregular and intersect in a larger region around $x = L/2$ for case 6 in Figure 7.11 (b).

Figure 7.11 Spatial variations of pressure along the symmetry axis of the enclosure at four different instants ($\omega t = 0, \pi/2, \pi, 3\pi/2$) during the 40th cycle: (a) case 5 and (b) case 6.
Figure 7.12 Spatial variations of $u$ velocity along the symmetry axis of the enclosure at four different instants ($\omega t = 0, \pi/2, \pi, 3\pi/2$) during the 40th cycle: (a) case 5 and (b) case 6.

The spatial variations of $u$ velocity along the symmetry axis of the enclosure at four different instants ($\omega t = 0, \pi/2, \pi, 3\pi/2$) during the 40th cycle: (a) case 5 and (b) case 6 are shown in Figure 7.12. The $u$ velocity profiles are distorted by the viscous and non-linear
effects for cases 5 and 6. A similar behavior for the u velocity along the symmetry axis is also observed as for the pressure profiles along the symmetry axis of the enclosure. Also, u velocity profiles in the enclosure are more sinusoidal for case 5 in Figure 7.12 (a), and they are more irregular for case 6 in Figure 7.12 (b).

Figure 7.13 Mean flow fields in the enclosure during the 40\textsuperscript{th} cycle: (a) case 5 and (b) case 6.

Figure 7.13 shows the mean flow fields in the enclosure during the 40\textsuperscript{th} cycle for cases 5 and 6. An initial state closer to the critical point does not affect the acoustic streaming
structure in the enclosure, nor do the enclosure height and maximum vibrating wall displacement. An interesting phenomenon is observed based on all the numerical results for cases with different initial states: the closer the initial state of the fluid in the enclosure, the less time to reach steady state secondary streaming flow field.

7.5 Conclusions

The formation of acoustic waves, acoustic-viscous boundary layer interactions, and associated flows in an enclosure filled with supercritical fluid nitrogen are numerically studied by solving the compressible Navier-Stokes equations in a two-dimensional Cartesian coordinate system. It is the first time that acoustic streaming is observed in supercritical fluids in numerical simulations.

The present study shows that no inner acoustic streaming is observed due to the extremely thin acoustic boundary layer thickness. Additionally, the initial state of the fluid in the enclosure, the maximum vibrating wall displacement and the ration of $H/\delta$, do not affect the acoustic streaming structure in the enclosure in the range $100 \leq X_{\text{max}} \leq 200$. The initial state of the fluid in the enclosure does not change the acoustic streaming structure in the enclosure, however it shortens the time to reach steady state secondary streaming flow field, as it is closer to the critical point.
CHAPTER 8
NUMERICAL INVESTIGATION OF HEAT AND MASS TRANSFER IN
SUPERCritical CARBON DIOXIDE

8.1 Introduction

Background discussion on heat and mass transfer in supercritical fluids has been
given earlier in Chapter 2 (Section 2.3). In the present chapter we present the results on the
phenomena of the natural convection heat and mass transfer in supercritical carbon dioxide.
For the heat transfer problem, a differentially heated square enclosure is considered.
Correlation for the steady state Nusselt number Nu as a function of heat transfer Rayleigh
number Ra_t (for a given Prandtl number Pr) are developed. For the mass transfer problem, a
heated naphthalene wall is considered, and a correlation is developed for the time-dependent
Sherwood numbers Sh(t) as a function of the mass transfer Rayleigh number Ra_m, and the
Fourier number Fo_m (for a given the Schmidt number Sc).

8.2 Problem Formulation

A square enclosure (Figure 8.1) filled with supercritical carbon dioxide is considered.
The dimensions of the enclosure are also given in Figure 8.1. For the heat transfer problem,
the left side wall is gradually heated to a given value while the right wall is maintained at a
constant temperature. The top and bottom wall are considered to be insulated. The direction
of gravity is as shown.
For the mass transfer problem, a heated naphthalene wall is considered. The naphthalene wall is gradually heated to a given value. The enclosure initially contains no dissolved naphthalene and the right wall is maintained at a constant temperature while the horizontal sides are thermally insulated.

### 8.2.1 Mathematical Model

The two dimensional compressible Navier-Stokes equations are considered.

- Conservation of mass

\[
\frac{\partial \rho}{\partial t} + \frac{\partial (\rho u)}{\partial x} + \frac{\partial (\rho v)}{\partial y} = 0
\]  \hspace{1cm} (8.1)

- Conservation of momentum in the x and y directions:

\[
\frac{\partial (\rho u)}{\partial t} + \frac{\partial (\rho uu)}{\partial x} + \frac{\partial (\rho uv)}{\partial y}
\]
\[
\frac{\partial p}{\partial x} + \frac{\partial}{\partial x} \left[ 2\mu \frac{\partial u}{\partial x} - \frac{2}{3} \mu \left( \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) \right] + \frac{\partial}{\partial y} \left[ \mu \left( \frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right) \right] = -\frac{\partial p}{\partial x} + \frac{\partial}{\partial x} \left[ \frac{\partial T}{\partial x} \right] + \frac{\partial}{\partial y} \left[ k \frac{\partial T}{\partial y} \right]
\]

(8.2a)

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

(8.2b)

- Conservation of energy

\[
\frac{\partial (\rho h)}{\partial t} + \frac{\partial (\rho u h)}{\partial x} + \frac{\partial (\rho v h)}{\partial y} = \frac{\partial p}{\partial t} + \frac{\partial}{\partial x} \left( k \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left( k \frac{\partial T}{\partial y} \right)
\]

\[
+ \frac{\partial}{\partial x} \left[ \mu \left( \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) \right] + \frac{\partial}{\partial y} \left[ 2\mu \frac{\partial v}{\partial y} - \frac{2}{3} \mu \left( \frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) \right]
\]

(8.3)

- Conservation of naphthalene concentration:

\[
\frac{\partial (C Y_{nap})}{\partial t} + \frac{\partial (C Y_{nap} u)}{\partial x} + \frac{\partial (C Y_{nap} v)}{\partial y} = -\frac{\partial}{\partial x} \left( -C D \frac{\partial Y_{nap}}{\partial x} \right) - \frac{\partial}{\partial y} \left( -C D \frac{\partial Y_{nap}}{\partial y} \right)
\]

(8.4)

where $Y_{nap}$ is the naphthalene mole fraction and

\[
C = \frac{\rho}{\left( Y_{nap} M_{nap} + \left(1 - Y_{nap}\right) M_{c02} \right)}
\]

(8.5)
8.2.2 Fluid Property

Properties of supercritical carbon dioxide are obtained from the NIST Standard Reference Database 12 [80] as functions of pressure and temperature. The fluid properties that are used in computations are density, viscosity, specific heat, enthalpy and thermal conductivity. The polynomial fits are used for generating fluid property equations from the database are given as:

\[ \rho = f(T,p), \quad \mu = f(T,p), \quad c_p = f(T,p), \quad h = f(T,p) \quad \text{and} \quad k = f(T,p). \]

8.2.3 Boundary Condition

The temperature at the left wall is gradually heated and the following expression is employed:

\[ T_i(t) = T_i + \Delta T \left[ 1 - \exp\left( -\frac{t}{\tau/c} \right) \right] \quad (8.6) \]

The values of \( \Delta T, \tau \) and \( c_1 \) are shown in Table 8.1. Here \( \Delta T \) is the intended temperature difference between the heated and the unheated wall, \( \tau = 2L/c \) where \( c \) is the acoustic speed in supercritical carbon dioxide and \( L \) is the width of the enclosure, and \( c_1 \) is a numerical constant that determines the rate of heating. The temperature at the right wall is maintained at the initial temperatures. The top and bottom walls are insulated. The enthalpies at the walls are a function of the temperatures and the pressures at the walls.

For the mass transfer problems considered, the Stefan velocity in the normal direction of naphthalene surface is given as:
Saturated mole fraction of naphthalene [67] at the left wall is considered ($Y_{\text{nap.sat}} = 0.0136$ at $T = 318.15$ K and $P = 11.8$ MPa) for the concentration of naphthalene at the left wall. The mass fluxes at the other walls are considered to be zero. Also no condensation of saturated naphthalene is considered along the right, top and the bottom walls. This assumption is justified as we simulate the mass transfer problem for a specified time (30 s), which is much shorter than the time required for the system to come to concentration equilibrium.

**8.2.4 Numerical Scheme**

A time-marching implicit finite volume method [93] was used to obtain the solutions to the governing equations with the specified boundary conditions. Euler 1st order differencing for time accuracy is chosen. For the convective-diffusive terms a second order upwind scheme [113] is used. The original idea of this second order upwind scheme is based on the evaluation of the cell face value by using linear interpolation of values at two upstream cells. The convergence criterion is 0.0001. Grid size for all cases is 100 by 100. Fine grids near the walls of the enclosure are applied. The ratio of maximum to minimum grid size is 5:1.

Because of gradual heating at the left wall, time step variation is also applied for the temporal calculations. Short time steps are used at the beginning of the process and the time steps are gradually increased as the flow develops. The time step variation is given as:

\[
\mathbf{u} = -M_{\text{nap}} \left( -CD \frac{\partial Y_{\text{nap}}}{\partial x} \bigg|_{x=0} \right) \rho_{\text{nap, solid}} \tag{8.7}
\]
\[ dt = dt_i + 0.1 \times \left[ 1 - \exp\left( -\frac{c_2 \tau}{\tau} \right) \right] \]  

(8.8)

where the initial time step \( dt_i = 10^{-6} \) s and the value of \( c_2 \) determines the rate of increase of the time step as the solution progresses. The CFD-ACE code is employed to solve the governing equations along with the initial and boundary conditions discussed earlier for the heat and mass problems in supercritical carbon dioxide.

Table 8.1 List of computational cases for the heat transfer studies

<table>
<thead>
<tr>
<th>Cases</th>
<th>Initial conditions</th>
<th>( \Delta T ) [K]</th>
<th>( \tau ) [s]</th>
<th>( c_1 )</th>
<th>( c_2 )</th>
<th>Gravity</th>
<th>Pr</th>
<th>( Ra_t )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>( T_i = 305 ) K</td>
<td>0.1</td>
<td>0.1</td>
<td></td>
<td></td>
<td>g</td>
<td></td>
<td>1.27 \times 10^9</td>
</tr>
<tr>
<td>2</td>
<td>( P_i = 7.5 ) MPa</td>
<td>1.17 \times 10^{-4}</td>
<td>0.01</td>
<td>0.001</td>
<td></td>
<td>0.1g</td>
<td>14.87</td>
<td>1.27 \times 10^6</td>
</tr>
<tr>
<td>3</td>
<td>0.1</td>
<td>0.1</td>
<td>0.01</td>
<td></td>
<td></td>
<td>0.01g</td>
<td></td>
<td>1.27 \times 10^7</td>
</tr>
<tr>
<td>4</td>
<td>0.1</td>
<td>0.1</td>
<td>0.001</td>
<td></td>
<td></td>
<td>0.001g</td>
<td></td>
<td>1.27 \times 10^6</td>
</tr>
</tbody>
</table>

8.3 Results and Discussions

The results are first presented for cases with heat transfer only (Table 8.1). The combined heat and mass transfer cases (cases 5 and 6) are discussed thereafter (Table 8.2). The value of the acceleration due to gravity is varied to obtain a range of \( Ra_t \) values as shown in Table 8.1.

The temperature contours in the enclosure at \( t = 30 \) s are shown in Figure 8.2 for all four cases. For the high \( Ra_t \) cases (Figs 8.2(a) and 8.2(b)), thin thermal boundary layers form along the vertical walls and the central region of the enclosure is essentially isothermal. For the system at \( Ra_t = 10^7 \) (Fig 8.2(c)), the temperature field reflects the effect of right-moving and left-moving flows along the top and the bottom sides. The effect of the weak flow field is not evident in Figure 8.2 (d) at \( Ra_t = 10^6 \).
Figure 8.2 Temperature contours in the enclosure at $t = 30$ s (a) case 1, (b) case 2, (c) case 3 and (d) case 4
Figure 8.3 Spatial local Nusselt numbers at the surfaces of the left and right at $t = 30$ s: (a) case 1, (b) case 2, (c) case 3 and (d) case 4

The spatial local Nu along the vertical surfaces of the enclosure at $t = 30$ s are shown in Figure 8.3. In all cases, high local Nu is observed along the top portion of the heated wall.
and the bottom portion of the unheated wall. It is interesting to note that in Figures 8.3(c) and 8.3(d), the maximum value of Nu occurs slightly below the top wall and slightly above the bottom wall. This is due to the formation of thick thermal boundary layers along the top and bottom walls as shown in Figures 8.2(c) and 8.2(d).

The time evolution of the spatially averaged Nu at the left and right walls of the enclosure is shown in Figure 8.4. The values at the left wall and the right wall converge to almost the same value in about 30 s. The slight differences are due to the slightly different thermal conductivity values of supercritical carbon dioxide along the two vertical walls. At the beginning, the temporal heat transfer rate at the left wall is high due to the heating. The value decreases until the system attains thermal equilibrium condition. For low Rat cases, the heat transfer along the right wall shows initial rise due to the so called ‘piston effect’

From the results, a correlation for the steady state Nusselt number at the heated wall is obtained as:

\[
\overline{Nu} = 0.1975 Ra^{0.2817} 
\]  

\[(8.9)\]

where \(1.27 \times 10^6 \leq Ra \leq 1.27 \times 10^9\) and \(Pr = 14.87\)

In past work, Catton’s correlation [114] for natural convection in enclosures is

\[
\overline{Nu} = 0.18 \left( \frac{Pr}{0.2 + Pr} Ra_L \right)^{0.29} \left[ \frac{1 \leq \frac{H}{L} \leq 2}{10^{-3} \leq Pr \leq 10^5} \right] \]

\[(8.10)\]
and H is the height of an enclosure and L is the width of the enclosure. If we substitute \( Pr = 14.87 \) and \( H = L \) in Equation 8.10, it yields

\[
\bar{Nu} = 0.18 Ra_L^{0.29}
\]

which is quite close to the correlation obtained in this study for supercritical carbon dioxide, Equation 8.9.

Now we present results for cases 5 and 6, where the left heated wall consists of naphthalene (see Table 8.2). The diffusion coefficient of naphthalene D in supercritical carbon dioxide for cases 5 and 6 is \( 1.356 \times 10^{-8} \) m\(^2\)/s [67].

For the mass transfer problem chosen, the steady state results are not obtained. In the steady state both the heat and mass transfers from the left wall and the right wall must balance. In the present formulation we do not consider any naphthalene (solid) deposition on the right, top and the bottom walls and hence the simulations are carried out for a specified length of time (\( Fo_m = 4.07 \times 10^{-3} \), \( t = 30 \) s). The mole fraction concentrations of naphthalene in supercritical carbon dioxide in the enclosure at \( t = 30 \) s are shown in Figure 8.5 for case 5 (\( Ra_m = 1.19 \times 10^9 \)) and case 6 (\( Ra_m = 1.19 \times 10^6 \)) at \( Fo_m = 4.07 \times 10^{-3} \). The high \( Ra_m \) case shows the formation of concentration boundary layers along the naphthalene wall and also vigorous dissolution of naphthalene in supercritical \( \text{CO}_2 \).
Figure 8.4 Temporal average Nusselt numbers at the left and right walls (a) case 1, (b) case 2, (c) case 3 and (d) case 4.
Table 8.2 List of computational cases for the mass transfer studies

<table>
<thead>
<tr>
<th>Cases</th>
<th>Initial conditions</th>
<th>Boundary conditions</th>
<th>( \tau ) (s)</th>
<th>( c_1 )</th>
<th>( c_2 )</th>
<th>Gravity</th>
<th>( Ra_m )</th>
<th>( Sc )</th>
<th>( Fo_m )</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>( T_i = 318.15 ) K</td>
<td>( T_L = \text{Eq. (8.6)} )</td>
<td>6.59 × 10^{-5}</td>
<td>0.01</td>
<td>0.001</td>
<td>1 g</td>
<td>1.19 × 10^{9}</td>
<td>5.59</td>
<td>4.07 × 10^{-3}</td>
</tr>
<tr>
<td>6</td>
<td>( P_i = 11.8 ) MPa</td>
<td>( T_R = 318.15 ) K ( (Y_N)_L = 0.0136 )</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

![Figure 8.5](image)

Figure 8.5 Mole fraction of naphthalene in supercritical carbon dioxide in the enclosure at \( t = 30 \) s (a) case 5 and (b) case 6

The local Sherwood numbers along the surfaces of the naphthalene at \( t = 30 \) s for cases 5 and 6 are shown in Figure 8.6. The local Sherwood numbers at the surfaces of the naphthalene at the lower level have the values higher than the local Sherwood numbers at the upper level.
The time evolution of spatially averaged Sh along the surface of the naphthalene slab is shown in Figure 8.7 for cases 5 and 6. The average Sh at the surface of the naphthalene slab have high values at the beginning time and decrease drastically as the concentration boundary layer starts developing. After that, the values decrease gradually. As expected, the system with high Ra has the Sh values higher than the system with the lower Ra.

It is noted that while the heat transfer problem reached steady state by about 30 s, the mass transfer problem will require a much longer time to reach steady state. We did not attempt to carry out the simulations of the mass transfer cases to steady state conditions and hence no naphthalene condensation was considered for the right wall.

From the results shown in Figure 8.7, a correlation for the transient spatially averaged Sh number at the surface of naphthalene was obtained:
\[ \overline{Sh} = 0.2099 R_{m}^{0.1855} F_{m}^{-0.2755} \]  

(8.11)

where \( 1.19 \times 10^6 \leq R_{m} \leq 1.19 \times 10^9 \), \( 0 \leq F_{m} \leq 4.07 \times 10^{-3} \) and \( Sc = 5.59 \).

8.4 Conclusion

Numerical simulations of heat and mass transfer in supercritical carbon dioxide are carried out for natural convection conditions in a differentially heated square enclosure. Properties of supercritical carbon dioxide are evaluated from the NIST Standard Reference Database 12 [80]. A correlation for the mean Nu along the heated wall is obtained as a function of the heat transfer \( Ra_{t} \) (for a given Prandtl number). The variation of \( Ra_{t} \) was primarily obtained by considering normal and reduced gravity conditions. The high \( Ra_{t} \) cases attain thermal equilibrium condition faster than the low \( Ra_{t} \) cases. For the heat and mass transfer cases, a correlation for the temporal \( Sh \) along the naphthalene surface was obtained.
as a function of the mass transfer $Ra_m$, and $Fo_m$ (for a given Schmidt number). The correlations agree well with available correlations found in the literature for gases and liquids.
CHAPTER 9
SUMMARY, CONCLUSIONS AND RECOMMENDATIONS

9.1 Summary

In this study, the mathematical model has been developed for handling the difference between the numerical simulations of ideal gases and supercritical fluids, due to the special property behaviors of supercritical fluids. These special properties include: equation of state, internal energy, viscosity, thermal conductivity and acoustic speed. Especially the equation of state and internal energy of a supercritical fluid exhibit a fundamental difference compared with the corresponding properties of an ideal gas. Unlike the linear equation of state for an ideal gas, the equation of state for a supercritical fluid exhibits strong non-linearity, especially in near-critical region. Also, the internal energy of a supercritical fluid is a non-linear function of both temperature and density (or pressure), while it is a linear function of temperature only for an ideal gas. These essential differences result in tremendous difficulties in numerical models (especially direct numerical simulations) for supercritical fluids.

The first problem is calculation for these special properties in numerical simulations for supercritical fluids. Table look-up method is discarded because of the large tables needed and the long computation time used for searching within tables. Two dimensional Polynomial fits are employed to calculate the properties of supercritical fluids as functions of temperature and density. A mathematical model has thus been developed to generate the polynomial fits via least square method, based on the data in the NIST Standard Reference
Database 12 [80]. The order of temperature and the order of density are adjusted according to the accuracy of the generated polynomial fits.

The second problem is the consistency of the method for calculating internal energy based on temperature and density, and the method for retrieving temperature based on internal energy and density. In direct numerical simulations for supercritical fluids, the internal energy of the fluid at initial state and the internal energy at boundaries after the initialization need to be determined based on the corresponding temperature and density. Also, the temperature of the fluid within the boundaries after initialization need to be retrieved based on the corresponding internal energy and density. In this study, the polynomial fit for internal energy as a function of temperature and density has been found to be sufficiently accurate when the order of temperature is second (In other words, the accuracy of the polynomial fit for internal energy is more sensitive to the order of density). Therefore a quadratic equation is derived from the polynomial fit based on known internal energy and density. After solving this quadratic equation, temperature is the positive root. Through this technique, the method for calculating internal energy based on temperature and density is consistent with that for retrieving temperature based on internal energy and density.

With the mathematical model mentioned above, the interactions between acoustic waves and supercritical fluids have been investigated numerically. Meanwhile, the physical mechanism of piston effect (fast thermal equilibration of supercritical fluids) is studied. The acoustic waves in supercritical fluids are generated by two kinds of means: rapid heating of the fluids, and mechanical vibrations of a solid wall. The temporal evolution of pressure,
density, temperature and flow field in supercritical fluids due to thermal and mechanical excitations are predicted. The model developed is also used to investigate the interaction of thermally induced acoustic waves and mass transfer in supercritical fluids (naphthalene dissolution in supercritical carbon dioxide).

9.2 General Conclusions

The generation, propagation and damping of thermally induced acoustic waves in supercritical nitrogen and carbon dioxide are studied by experimental and numerical approaches. In both experimental and numerical results, thermally induced acoustic waves are generated by rapid heating of the enclosure wall, and the strength of the pressure waves has been found to be proportional to the wall heating rate. In the acoustic time-scale, thermally induced acoustic waves play an important role for piston effect. The cause of piston effect in the intermediate time scale is explained by the fact that the internal energy of a supercritical fluid is a function of temperature and density (or pressure).

The flow fields and heat transport induced by thermally induced acoustic waves and buoyancy in an enclosure filled with supercritical carbon dioxide are investigated numerically. At early time, thermally induced acoustic waves are found to play an important role in temperature field and flow filed, enhancing heat transfer and fluid flow in the enclosure. The thermally induced acoustic waves also result in the highly oscillatory nature of the wall heat fluxes at early time. As the acoustic waves damp out, buoyancy-induced natural convection vortices begin to dominate the flow field. It has also been found that the buoyancy induced flow fields and natural convection are both affected by the initial state of
the fluid in the enclosure. This phenomenon can be explained by the higher Grashof number due to the lower kinematic viscosity, when the initial state is closer to the critical point.

Numerical simulations of heat and mass transfer in a differentially heated enclosure filled with supercritical carbon dioxide are carried out. A correlation for the mean Nu along the heated wall is obtained as a function of the heat transfer $Ra_t$ (for a given Prandtl number). The variation of $Ra_t$ was primarily obtained by considering normal and reduced gravity conditions. The high $Ra_t$ cases attain thermal equilibrium condition faster than the low $Ra_t$ cases. For the heat and mass transfer coupled cases, expressions for temporal Sh along the naphthalene surface is obtained as a function of the mass transfer $Ra_m$, and $Fo_m$ (for a given Schmidt number). The expressions exhibit a good match with the available ones in the literature for gases and liquids.

The formation of acoustic waves, acoustic-viscous boundary layer interactions, and associated flows in an enclosure filled with supercritical fluid nitrogen are numerically studied. It is the first time that acoustic steaming is numerically observed in an enclosure filled with a supercritical fluid. It has been found that the initial state of the fluid in the enclosure, the maximum vibrating wall displacement and the ration of $H/\delta_v$ do not affect the acoustic streaming structure in the enclosure in the range $100 \leq X_{max} \leq 200$. The present study also shows that no inner acoustic streaming is observed due to the extremely thin acoustic boundary layer thickness. Additionally, the initial state of the fluid in the enclosure does not change the acoustic streaming structure in the enclosure, however it shortens the time to reach steady state secondary streaming flow field, as it is closer to the critical point.
9.3 Recommendations for future work

Supercritical fluids are widely used in various industrial and laboratory processes. Similarly, acoustic-fluid dynamic interactions also have a wide range of industrial applications. In order to fully explore their underlying physics, the following research tasks are recommended.

9.3.1 Extension of numerical model to two phase zone

The present numerical models cannot study flow fields and heat transport in near-critical two phase zone. Thus for current numerical simulations, the initial states of the supercritical fluid are away enough from the critical point to avoid two phase zone. Therefore these models need to be extended to handle the near-critical two phase zone. The important applications of the extended models includes the simulations of acoustic streaming and thermally induced acoustic waves in supercritical fluids, whose states are extremely close to the critical point or in the near-critical liquid zone.

9.3.2 Soret effect and Soret coefficient in binary supercritical mixtures

Soret effect was discovered by the Swiss scientist Charles Soret in 1879 [115]. He observed that a salt solution contained in a tube with the two ends at different temperatures did not remain uniform in composition. This phenomenon, in which the temperature gradient induces a mass flux, was named as Soret effect. Considering Soret effect, the x component of the mass flux of a component in a binary mixture is [116]:

\[ J_x = -\rho D \frac{\partial c}{\partial x} - \rho D_T c_0 (1 - c_0) \frac{\partial T}{\partial x} \]

where the first term in the right hand side of above equation is Fick’s law of diffusion. C is
the mass fraction of the species and D is the molecular diffusion coefficient. The second term in the right hand side describes the Soret effect, with \( D_T \) the thermo-diffusion coefficient. The ratio of \( D_T/D \) is defined as Soret coefficient.

As heat and mass transfer in supercritical mixtures are attracting researching interests, because supercritical fluids are widely used in various industrial and laboratory processes as substitutes for organic solvents, Soret effect and Soret coefficient show more and more importance. So far Nakano is the only researcher who numerically proved the existence of Soret effect in supercritical binary mixtures and determined the sing of Soret coefficient, based on a simplified model. However detailed characterization of Soret effect and the exact values of Soret coefficient for supercritical binary mixtures are still unexplored.

**9.3.3 Modeling mass transfer in trans-critical zone**

The present numerical models cannot study heat and mass transfer in near-critical two phase zone. With an extended numerical model which can handle near-critical two phase zone, mass transfer in trans-critical zone will be of great importance and interests, due to the existence of surface tension in sub-critical zone and the disappear of surface tension in supercritical zone. This phenomenon is caused by the fact that liquid/gas phase boundaries exist in sub-critical fluids, while there is no liquid/gas phase boundary in supercritical fluids. When the critical point is transcended, many thermophysical properties exhibit interesting and special changes. These special behaviors of thermophysical property and the surface tension near the point will result in interesting mass transfer problems.
9.3.4 Modeling of Mass transfer enhancement in supercritical fluids by means of mechanically driven acoustic waves

The use of mechanically driven acoustic waves, especially ultrasound waves, has been increased during recent years. The mechanically driven acoustic waves represent a potential efficient way of enhancing mass in numerous supercritical fluids extraction processes [72]. Although some experimental work has been conducted in this area, the interaction of mass transfer and mechanically driven acoustic waves in supercritical fluids is still not fully studied. The present model can be extended to model Mass transfer enhancement in supercritical fluids by means of mechanically driven acoustic waves.

9.3.5 Parallelization improvement of codes

The codes of present model are quite time-consuming, due to the large mesh size and the high order two dimensional polynomial fits needed. The large mesh size and the high order two dimensional polynomial fits are caused by the strong non-linearity of thermophysical properties, especially the equation of state and internal energy, in near-critical region. All the simulations in this study were conducted in serial computers. The codes can thus be parallelized to significantly reduce the computation time.
APPENDIX A: NOMENCLATURE

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>constant controlling heating rate</td>
</tr>
<tr>
<td>c</td>
<td>speed of sound</td>
</tr>
<tr>
<td>cp</td>
<td>specific heat at constant pressure</td>
</tr>
<tr>
<td>cv</td>
<td>specific heat at constant volume</td>
</tr>
<tr>
<td>D</td>
<td>molecular diffusion coefficient</td>
</tr>
<tr>
<td>Dr</td>
<td>thermo-diffusion coefficient</td>
</tr>
<tr>
<td>i</td>
<td>internal energy</td>
</tr>
<tr>
<td>E</td>
<td>total energy</td>
</tr>
<tr>
<td>f</td>
<td>frequency</td>
</tr>
<tr>
<td>Fo_m</td>
<td>mass transfer Fourier number</td>
</tr>
<tr>
<td>g</td>
<td>gravitational acceleration</td>
</tr>
<tr>
<td>H</td>
<td>enclosure height</td>
</tr>
<tr>
<td>h</td>
<td>heat transfer coefficient</td>
</tr>
<tr>
<td>k</td>
<td>thermal conductivity</td>
</tr>
<tr>
<td>L</td>
<td>enclosure width</td>
</tr>
<tr>
<td>Nu</td>
<td>Nusselt number</td>
</tr>
<tr>
<td>Ra_t</td>
<td>Rayleigh number</td>
</tr>
<tr>
<td>Ra_m</td>
<td>mass transfer Rayleigh number</td>
</tr>
<tr>
<td>p</td>
<td>pressure</td>
</tr>
<tr>
<td>q</td>
<td>heat flux</td>
</tr>
<tr>
<td>R</td>
<td>specific gas constant</td>
</tr>
</tbody>
</table>
Ra  Rayleigh number
Ra_m  Mass transfer Rayleigh number
Sh  Sherwood number
t  time
T  temperature
u  velocity component in the horizontal direction
v  velocity component in the vertical direction
X  maximum wall displacement
x  horizontal coordinate
y  vertical coordinate

Greek symbols
α  thermal diffusivity
β  volumetric thermal expansion coefficient
δν  acoustic boundary layer thickness
λ  wavelength
µ  dynamic viscosity
ν  kinematic viscosity
ρ  density
τ  travel time for the acoustic wave to traverse the enclosure width
ω  angular frequency

Subscripts and superscripts
L left
n direction normal to the wall
R right
APPENDIX B: TWO DIMENSIONAL POLYNOMIAL FITS

The general format of two dimensional polynomial fits is:

\[ p = \sum_{i=0}^{n1} \sum_{j=0}^{n2} c_{ij} (T - T_{ref})^i (\rho - \rho_{ref})^j + p_{ref} \]

Following two dimensional polynomial fit is for pressure as a function of temperature and density for supercritical carbon dioxide within this range: 310 K < T < 312 K and 7.8 MPa < p < 8.1 MPa.

\( T_{ref} = 311 \text{ K}, \rho_{ref} = 325 \text{ Kg/m}^3, p_{ref} = 7.95 \text{ MPa}, n1 = 6, n2 = 6 \)

Table B.1 Coefficients of a polynomial fit for pressure as a function of temperature and density for supercritical carbon dioxide (Part I: 0 \leq i \leq 3)

<table>
<thead>
<tr>
<th>C_{ij}</th>
<th>i</th>
<th>0</th>
<th>1</th>
<th>2</th>
<th>3</th>
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</thead>
<tbody>
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<td>0</td>
<td></td>
<td>151159.1876133448</td>
<td>113357.690743503</td>
<td>-244.001947887921</td>
<td>-22.8207370780922</td>
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<td>5080.62608314034</td>
<td>429.462231002512</td>
<td>-4.63257507027590</td>
<td>-5.22754177314049</td>
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</tr>
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<td>-23.0367984666202</td>
<td>-0.274037258515608</td>
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<td>-1.823606361401806E-2</td>
<td>-1.834611541515606E-2</td>
<td>-3.548837616481414E-4</td>
<td></td>
</tr>
<tr>
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<td>1.211280437957571E-4</td>
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<td>5</td>
<td>-4.762130959958319E-6</td>
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<td>1.532942754618008E-6</td>
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<tr>
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<td>-3.186707234588157E-8</td>
<td>-2.451318686005516E-8</td>
<td>7.474638254698547E-9</td>
<td>1.263069893844311E-9</td>
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</tr>
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</table>
Table B.2 Coefficients of a polynomial fit for pressure as a function of temperature and density for supercritical carbon dioxide (Part II: $4 \leq i \leq 6$)

<table>
<thead>
<tr>
<th>$C_{ij}$</th>
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<th>5</th>
<th>6</th>
</tr>
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<tbody>
<tr>
<td>0</td>
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<td>0.169566135643654</td>
<td>4.13440521504119</td>
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<tr>
<td>1</td>
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<td>0.272729939138776</td>
<td>2.633278891879676E-2</td>
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<td>2</td>
<td>6.189716087937557E-2</td>
<td>-1.395961880058124E-3</td>
<td>-2.856462131255849E-2</td>
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<tr>
<td>3</td>
<td>4.101854674682688E-3</td>
<td>-1.283184617323215E-3</td>
<td>-1.082171921769529E-3</td>
</tr>
<tr>
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<td>4.360467007719197E-5</td>
<td>-3.747527935248842E-5</td>
<td>-8.664957586425404E-6</td>
</tr>
<tr>
<td>5</td>
<td>-1.941798538801024E-7</td>
<td>-2.103940332774207E-7</td>
<td>3.468509082564806E-8</td>
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<tr>
<td>6</td>
<td>-1.576868782466711E-9</td>
<td>8.610850855754752E-10</td>
<td>2.18990153011337E-10</td>
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</tbody>
</table>
APPENDIX C: LIST OF PUBLICATIONS

The present research study resulted in a number of scientific publications. A list of the journal articles and the conference publications is given below (including the papers under preparation):

Journal Publications


- Z. lei, B. Farouk, Natural Convection in an Enclosure filled with Supercritical Carbon Dioxide, Journal of Thermophysics and Heat Transfer (in preparation)

Conference Publications:

- Z. Lei, B. Farouk, Convection and Transport in a Differentially Heated Enclosure Filled with Supercritical Carbon Dioxide: Short- and Long-Time Solutions, ASME IMECE, Boston, MA, 2008

- U. Prasopchingchana, Z. Lei, B. Farouk, Heat and Mass Transfer in a Differentially Heated Enclosure Filled with Supercritical Carbon Dioxide, ASME IMECE, Boston, MA, 2008

- Z. Lei, B. Farouk, Generation and Propagation of Thermally Induced Acoustic Waves in Supercritical Carbon Dioxide, ASME IMECE, Seattle, WA , 2007


- Z. Lei, B. Farouk, Generation and Propagation of Thermoacoustic Waves in Mildly Supercritical Carbon Dioxide, ASME Heat Transfer Division, 2006

- Z. Lei, B. Farouk, Numerical Simulation of 'Piston Effect' in Supercritical Carbon Dioxide, Heat Transfer Summer Conference, San Francisco, CA, 2005
- Z. Lei, et al., Generation and Propagation of Thermoacoustic Waves in Mildly Supercritical Carbon Dioxide, ASME International Mechanical Engineering Congress, Anaheim, California, 2004
Zhiheng Lei was born in Sichuan Province, P. R. China on May 18, 1975. He received his B.S. degree at Hebei Institute of Architecture and Civil Engineering, China in 1997, and his M.S. degree at Tianjin University, China in 2000. He joined the doctoral program in the Department of Mechanical Engineering and Mechanics at Drexel University, Philadelphia, Pennsylvania, USA in 2004. He has been a research assistant and a teaching assistant through his years at Drexel University. His research interests include the computational and the experimental investigations of fluid dynamics and heat transfer problems in supercritical fluids.
REFERENCES


