FLORIDA STATE UNIVERSITY

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THIS IS MY TITLE:

AND THIS IS ITS SECOND LINE

By

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To my parents, Jeff and Maria.

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I'd like to thank the academy...

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LIST OF ABBREVIATIONS

The following short list of abbreviations are used throughout this document.

AMR	Adaptive Mesh Refinement
CFD	Computational Fluid Dynamics
CO2	Carbon Dioxide
DNS	Direct Numerical Simulation
DOE	Department of Energy
ECP	Exascale Computing Project
EoS	equation of state
HPC	high-performance computing
LES	Large Eddy Simulation
NIST	National Institute of Standards and Technology
NREL	National Renewable Energy Laboratory
PPM	piecewise parabolic method
PR EoS	Peng-Robinson equation of state
RANS	Reynolds-Averaged Navier-Stokes
rms	root mean square
sCO2	Supercritical Carbon Dioxide
SGS	Subgrid Scale
SMD	dynamic Smagorinsky
SRK EoS	Soave-Redlich-Kwong equation of state

ABSTRACT

This is my abstract.

CHAPTER 1

INTRODUCTION

Fluids are a vital part of everyday life - two of the most common ones being the air we breath and the water we drink. Less obvious are all the ways even these common fluids function behind the scenes in our day to day activities. Water, for example, serves an important role in various industrial settings, proving useful in areas ranging from thermal management to energy production; steam turbines alone accounted for 45% of electricity generation in the United States in 2021 [16]. Improvements to these types of systems impact technologies across a variety of fields and are thus an important area of research. One strategy toward this end, and the primary motivator of the work presented here, is the use of supercritical fluids as the working fluid of these systems.

In order to discuss supercritical fluids further, we must first go into more detail about general fluids. A *fluid* is a large collection of mutually interacting particles (e.g. molecules, atoms, etc.) in a state of constant and chaotic motion. This results in the continuous deformation of the substance under the effects of a shearing stress. The two categories of fluids that most people are familiar with are gases and liquids. *Liquids* are (mostly) incompressible and have definite volume for a set temperature and pressure. *Gases* are compressible and do not have definite volume. A simplified distinction between the two is that both gases and liquids will conform to the shape of whatever container they are in, but gases will further spread to fill all available space present.

A *supercritical fluid* is a fluid that is held above a critical temperature and pressure, at which point the distinction between a gas and liquid phase no longer exists [4, 17]. Supercritical fluids have qualities associated with both gases and liquids yet simultaneously have features that exclude them from fully being categorized as one or the other. For example, while they have viscosities akin to gases, they have solvent capabilities associated with liquids []. Similarly, while they have densities in line with liquids, they lack surface tension []. One benefit of this duality is that supercritical fluids can be fine tuned to be more gas-like or more liquid-like depending on the application at hand. This also results in ambiguity on how to actually classify them, with some sources considering them highly compressed gases [6], expanded liquids [1], or even as their own distinctly separate phase [2]. The distinction usually lies on the specifics of the regime and the application at hand.

This work is concerned with supercritical Carbon Dioxide (sCO2) in particular. sCO2 has many beneficial features to a wide variety of industrial applications, as we will detail further in section []. Many of the applications of interest to this work involve injection technologies that require a round turbulent jet configuration within the system. While much research has gone into sCO2 flows, the current landscape is lacking in turbulence of this type with the context of understanding the underlying physics relating to the flow itself. For research that does involve other supercritical turbulent jets, the regimes explored in those works typically involve transcritical fluid injection within the regime of interest or explore regions outside the scope of this work.

The goal of this work is to explore the pseudo-boiling region of the pseudo-critical zone and analyze the influence of extreme thermodynamic fluctuations on turbulence statistics within the flow field. To that end, the rest of this chapter continues as follows: first, the mathematical framework for modeling compressible Newtonian fluids is provided to form the basis of the modeling done in this dissertation. Further consideration is then given to turbulence modeling and the numerical methods developed for studying turbulence to provide insight into the quantities of interest analyzed within this dissertation and the choices of numerical methods used herein. Important applications of supercritical carbon dioxide in particular are provided to motivate the problem presented in this dissertation. Existing numerical studies on supercritical fluids are reviewed to demonstrate how this dissertation fits into the current landscape of research and to emphasize the contribution the results of this work make to the field. This chapter concludes with an outline of the dissertation, the goals of the dissertation, and the main contributions made through this work.

1.1 Mathematics of Fluid Flow

Scale is one of the key factors to consider when developing a mathematical description of a fluid system. For example, consider modeling flow past a satellite in the exosphere vs. the flow past a turtle in the ocean; these two mediums have vastly different characteristics and would thus require different modeling techniques. Scale is also an important concept when it comes to turbulence in particular so we will begin that discussion here with our choice in perspective for the mathematical framework of our system of interest.

From a kinetics perspective, particle motion within a fluid can be broken up into two phases: particle interaction and free flight. Average time spent in free flight, $< t_f >$, is typically much greater than collision time for a given interaction, t_c. The average length traveled between collisions

is known as the mean free path, ℓ. Since free flight time dominates particle interaction time, this phase determines the length scale of the kinetic description of motion. In addition to this inherent physical scale, there is also a scale associated with the resolution of the problem itself, L. These two scales are important, as the mathematical description of your model depends on how these two scales compare to one another. This comparison is related through the non-dimensional Knudsen number:

$$Kn = \frac{\ell}{L} \tag{1.1}$$

Flows with large Knudsen number ($Kn \gg 10$) require modeling from the kinetic or microscopic perspective as particle interactions become sparse enough compared to the scope of the problem to require a statistical mechanics framework. On the other hand, small Knudsen number flows ($Kn \ll 0.01$) have a problem scale that far exceeds the particle-level interactions present, giving way to an average overall motion within the fluid. In these flows, a continuum description of the fluid is appropriate for capturing this macroscopic behavior.
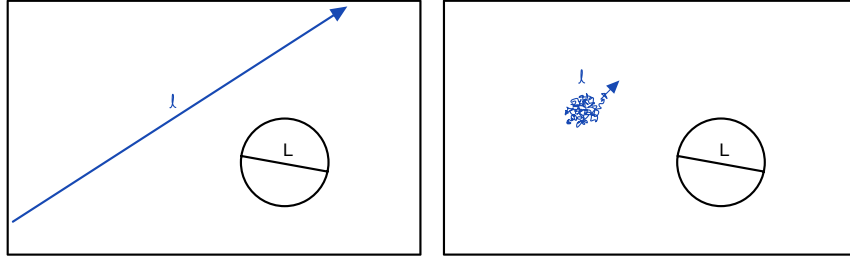
Figure 1.1: Characteristic length scale of problem, L, compared to mean free path of particles, ℓ, for a flow with large Knudsen number (left) vs. small Knudsen number (right)

This work falls within the small Knudsen regime, so we will be working with the continuum description of fluids. In this section we will discuss the Navier-Stokes Equations that arise from this modeling technique and how we account for the supercritical nature of the flow through our choice of equation of state.

1.1.1 Continuum Description of Fluids

The continuum hypothesis assumes that the fluid has no fine structures and that it is perfectly continuous, i.e. the properties of a small subdivision are the same as other subdivisions. This allows for the approximation of physical quantities at the infinitesimal limit [7].

For example, consider a fluid with arbitrary volume V as depicted in Figure 1.2.
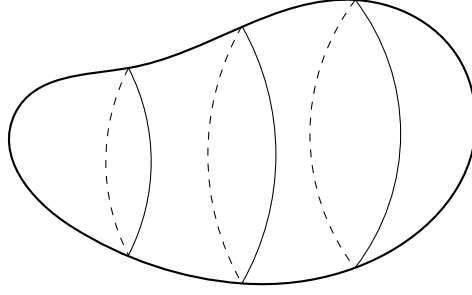
Figure 1.2: A fluid of arbitrary volume V bounded by surface S with velocity $\vec{v}(\vec{x}, t)$. A differential volume and surface area is given by dv and ds, respectively. $\vec{n}$ is the outward-pointing unit normal vector to the surface S.

For a fluid with density $\rho(\vec{x}, t)$, mass within a small representative volume can be described with

$$\rho\, dv$$

Total mass in the arbitrary volume is then given by

$$\iiint_V \rho\, dv$$

The rate of change of mass through the volume is now

$$\frac{d}{dt} \iiint_V \rho\, dv$$

$$= \iiint_V \frac{\partial \rho}{\partial t}\, dv \tag{1.2}$$

Simultaneously, overall change in mass throughout the volume can be described by the net mass flux through the surface S. Volumetric flow through a small portion of the bounding surface is given by

$$\vec{v} \cdot \vec{n}\, ds$$

Total mass flux through the entire surface is then

$$\iint_S \rho\, \vec{v} \cdot \vec{n}\, ds \tag{1.3}$$

Applying the divergence theorem to Eq. 1.3 yields the following volume integral

$$\iiint_V \nabla \cdot (\rho\, \vec{v})\, dv \tag{1.4}$$

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Assuming there is no additional source generating or leaking mass within the control volume, we can relate Eqs. 1.2 to 1.4 :

$$\iiint_V \frac{\partial \rho}{\partial t} \, dv = -\iiint_V \nabla \cdot (\rho \, \vec{v}) \, dv$$

$$\iiint_V \frac{\partial \rho}{\partial t} \, dv + \iiint_V \nabla \cdot (\rho \, \vec{v}) \, dv = 0$$

$$\iiint_V \left(\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \, \vec{v}) \right) dv = 0 \tag{1.5}$$

Note the inclusion of the negative sign for the right side of the initial equality; in the surface integral formulation, the outward facing normal describes flux out of the volume, thus yielding a decrease in mass within the volume. Since Eq. 1.5 holds for any arbitrary volume V, the integrand must be identically equal to zero:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \, \vec{v}) = 0 \tag{1.6}$$

Through the continuum hypothesis and conservation of mass, we have now arrived at the continuity equation in Eq. 1.6. This specific process demonstrates an even more fundamental relationship known as a *conservation law*. More generally, for some integrated property ϕ, the rate of change of ϕ within a control volume must be equal to the amount of ϕ lost or gained through the boundaries of the control volume plus what is created or consumed by any sinks or sources, s, within the volume (sinks having positive orientation to match the positive orientation of the outward-facing normal $\vec{n}$) [].

$$\frac{\partial \phi}{\partial t} + \nabla \cdot (\phi \, \vec{v}) + s = 0 \tag{1.7}$$

In addition to this concept applying to conservation of mass, as was seen in this section, the idea outlined by Eq. 1.7 applies to conservation of momentum and energy within the fluid. Together, these expressions combine to form the basis of the Navier-Stokes Equations, as will be seen in more detail in chapter 2. The important takeaway from this section is that with the continuum hypothesis and fundamental laws of physics, one can adequately capture macroscopic flow behavior for the types of flows we are interested in within this work.

1.1.2 Equation of State

Conservation of mass, momentum, and energy gives us five equations to describe our fluid system. For compressible flows, this is not enough information to solve for all the unknowns within the system of coupled partial differential equations. A sixth equation, known as the Equation

of State (EoS), must be chosen in order to close the system. The EoS relates three of the six unknowns: pressure, temperature, and density. Here we will briefly discuss some EoS options and their distinguishing characteristics in order to motivate the choice made for this work.

The simplest option available is the ideal gas EoS, which comes from the ideal gas law. This EoS relates density, pressure, and temperature in the following manner:

$$p = \frac{RT}{V_m} \tag{1.8}$$

where p is pressure, R is the universal gas constant, T is temperature, and $V_m = \frac{V}{n}$ is the molar volume of the fluid (it is common to express density in terms of molar volume for sake of simplicity in writing the EoS with V being volume and n being the number of moles). The ideal gas EoS is fairly accurate for liquids and gases at moderate temperatures and low pressures. It fails at low temperatures and high pressures, especially near the transition region from gas to liquid. The inaccuracy noted in this region means this EoS would not be suitable for the area of interest within this study.

Cubic EoS generally provide more accuracy than the ideal gas EoS. The first cubic EoS was developed by van der Waal in 1873, modifying the ideal gas EoS to take into consideration the finite size of molecules and interactions between molecules (the ideal gas EoS only accounts for interactions with the container and treats molecules as point particles). Other cubic EoS can be thought of as modifications from this base form:

$$p = \frac{RT}{V_m - b} - \frac{a}{V_m^2} \tag{1.9}$$

where a and b are constants related to the pressure and temperature at the critical point, p_c and T_c respectively:

$$a = \frac{27(RT_c)^2}{64p_c}, \quad b = \frac{RT_c}{8p_c}$$

One of the main benefits of using a cubic EoS is that they can have comparable and sometimes even better accuracy compared to their higher order counterparts, thus reducing computational costs. However, it is important to take into consideration the regime of interest in addition to the fluid of interest when choosing an EoS, as each one has its own pros and cons. For example, molecule polarity and density are two factors that can have a high impact in selection between the Soave-Redlich-Kwong Equation of State (SRK EoS) and Peng-Robinson Equation of State (PR EoS) alone [5].

This work uses the SRK EoS as will be more thoroughly introduced in chapter 2. It has been shown that this equation of state is fairly accurate for the parameter regime under careful consideration in this work []. This accuracy is detailed further in chapter 3 through comparisons with data from National Institute of Standards and Technology (NIST). Overall, when adequately considered, the EoS is the key avenue to incorporating specific fluid properties into the mathematical model.

1.2   Turbulence

Additionally, fluid flow can be categorized into different types based on certain defining flow characteristics. The main two classifications of note are laminar flow and turbulent flow.

Laminar flow is denoted by fluid particles having well-defined parallel trajectories of motion, or streamlines. Streamlines do not cross, meaning adjacent layers within the fluid flow by one another with little to no mixing. From a more generalized perspective, the flow appears to be smooth. In contrast to this, turbulent flow is characterized by its unpredictable and chaotic trajectories. Streamlines do cross resulting in swirls and eddies of varying length scales which induce mixing. Turbulent flow can be qualitatively described as being rough due to this high degree of fluctuation within the velocity and pressure fields present. This generalized description is depicted in Figure 1.3.
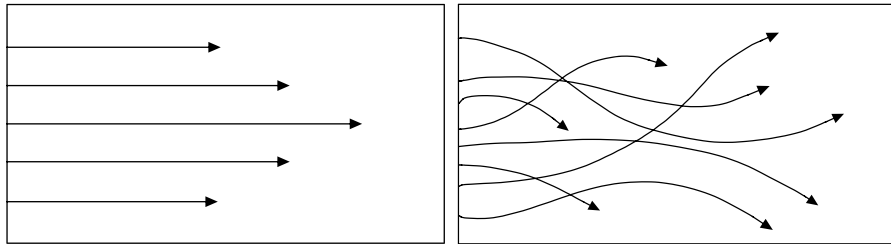
Figure 1.3: Example of streamlines in laminar (left) vs. turbulent (right) flow.

The Reynolds number is a dimensionless value that can be used to distinguish laminar flow from turbulent flow. It is defined as follows:

$$\mathrm{Re} = \frac{\rho u L}{\mu} \tag{1.10}$$

where ρ and μ are the density and dynamic viscosity of the fluid, respectively, u is the flow velocity, and L is a characteristic length scale associated with the given flow scenario (e.g. pipe diameter).

As is demonstrated by the ratio in Equation 1.10, the Reynolds number measures the relative effects of inertial forces compared to viscous forces within a given flow scenario. A small Reynolds number signifies the dominance of viscous forces; fluid parcels moving in tandem want to "stick together," resulting in the sheared flow and parallel trajectories seen in laminar flow. Turbulence is then characterized by a large Reynolds number, where inertial forces take precedence. Here, deviations within the laminar flow field result in lateral mixing between shear layers. This creates eddies and random trajectories that result in the chaotic motion of turbulent flow.

This work focuses on the turbulent round jet and its associated dynamics in the context of supercritical fluids. The remainder of this section details a brief historical overview of turbulence modeling and numerical methods developed for studying turbulence in order to motivate the modeling and numerical choices made within this work.

1.2.1 Historical Perspective

1.2.2 Numerical Approaches

1.3 Supercritical Carbon Dioxide

As mentioned earlier, supercritical fluids have many qualities that make them desirable as working fluids in a variety of systems. We now shift our focus to one particular fluid of interest: Carbon Dioxide (CO2). As seen in the phase diagram of Fig. 1.4, the critical temperature, T_c, and critical pressure, p_c, of CO2 are 304.128 K and 73.773 bar. The critical temperature and pressure of CO2 is fairly easy to attain, making it a strong candidate for systems with hight thermal outputs. Additionally, sCO2 has a relatively low toxicity and environmental impact [], and is chemically stable, non-flammable, and readily available []. For these reasons, sCO2 is a highly coveted alternative working fluid in many different applications, and is one of the most widely used supercritical fluids along with water [4].

In the next part of this section, we will explore some applications of sCO2 in which the turbulent round jet configuration is used. Then we will go through a brief review of recent studies involving sCO2, and more generally supercritical turbulent jets, in order to demonstrate where this work fits in among current research. Finally, we outline the structure of the remainder of the dissertation.
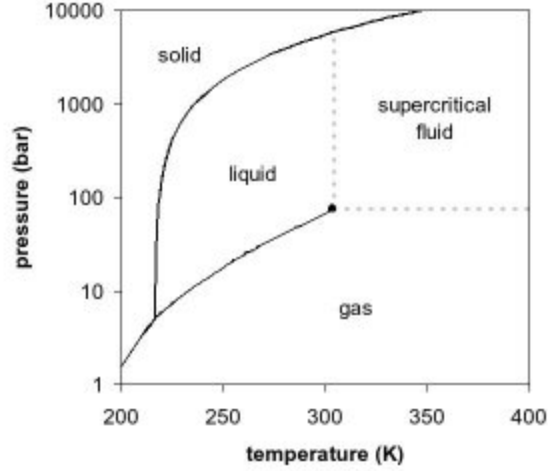
Figure 1.4: Phase diagram for Carbon Dioxide (CO2). Critical pressure, p_c, and temperature, T_c, are 73.773 bar and 304.128 K, respectively.

1.3.1 Applications of Interest

1.3.2 Overview of Current Numerical Landscape

Much research has gone into the development of appropriate numerical methods for such investigations, though challenges arise due to the lack of experimental data for use in model validation. Studies using direct numerical simulation (DNS) have been implemented to help establish benchmark test cases for other types of numerical schemes. Ruiz et al. use 2D DNS to simulate a mixing layer created by two streams of supercritical Oxygen and gaseous Nitrogen, using two different computational fluid dynamics (CFD) solvers to add confidence to their results [13]. A 3D DNS is used by Ries at al. to simulate a round Nitrogen jet for comparison with experimental data produced by Mayer et al. [12]. However, this study requires a reduction in Reynolds number from 1.62×10^5, based on the injection diameter, to 5300 in order to feasibly execute the computations. Li also utilizes a low Reynolds number of 1750 to study a round turbulent sCO2 jet with a preconditioning scheme [8]. The Reynolds-averaged Navier-Stokes (RANS) approach has also been implemented utilizing theory from the ideal gas case [9], but with the goal of ascertaining a more general understanding of why specifically sCO2's rock-breaking ability is better than that of water. In order to maintain a high Reynolds flow and better capture the effects of the supercritical nature of the fluid on the turbulence dynamics, the use of large eddy simulation (LES) has been explored. The impact of subgrid scale (SGS) models in capturing transcritical and supercritical dynamics

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of cryogenic Nitrogen have been analyzed through comparison with the Mayer et al. experiment and highly accurate NIST data [11, 19, 20, 10, 18]. Schmitt et al. does a similar investigation using LES, then extending their investigation to include sCO2 after validation with the Mayer et al. data. [14] However, this investigation uses low-pressure jets and does note the SGS models might need additional contributions to handle non-linearities and the pressure regime. While many of these investigations note that SGS models may need modification to deal with supercritical flows [14, 11, 10, 19], it is noted by Muller et al. that given a sufficiently fine grid, the influence of SGS modeling and numerical flux discretization is essentially limited to second-order moments [10]. Thus, we will be using the compressible version of the dynamic Smagorinksy SGS closures for our investigation, with further consideration of any influence of SGS model on our quantities of interest being noted later on. While much of the literature thus far has explored the impact of different numerical methods on modeling supercritical fluid flows and has aimed to strengthen the validity of these simulations in spite of the lack of experimental data available in the current landscape, a general consensus has still not been reached on how the supercritical nature of these fluids impacts the turbulence physics of these models. Thus, there remain open questions for understanding the fundamental flow behavior of turbulent jets in a supercritical environment, especially near the supercritical point, where both experimental and numerical investigations are still a challenge.

CHAPTER 2

MODEL OVERVIEW

2.1   Introduction

Chapter 1 introduced the general mathematical framework needed for modeling fluid flow.

2.2   Governing Equations

We consider the three-dimensional compressible Navier-Stokes equations:

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_j}\left(\rho u_j\right) = 0,$$

$$\frac{\partial}{\partial t}\left(\rho u_i\right) + \frac{\partial}{\partial x_j}\left(\rho u_i u_j + p\delta_{ij} - \sigma_{ij}\right) = 0, \tag{2.1}$$

$$\frac{\partial}{\partial t}\left(\rho E\right) + \frac{\partial}{\partial x_j}\Big(\left(\rho E + p\right)u_j + q_j - \sigma_{ij}u_i\Big) = 0$$

where ρ is the density, u_j is the velocity for the x_j direction, p is the pressure, $E = e + \frac{u_i u_i}{2}$ is the total energy, $e = c_v T$ is the internal energy, T is the temperature, and c_v is the heat capacity at constant volume. Additionally, the diffusive fluxes are

$$\sigma_{ij} = 2\mu S_{ij} - \frac{2}{3}\mu \delta_{ij} S_{kk} q_j = -k\frac{\partial T}{\partial x_j} \tag{2.2}$$

where $S_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right)$ is the strain-rate tensor, μ is the dynamic viscosity, and k is the thermal conductivity. External forces such as gravity are not included in this study. Chung's high pressure correction for viscosity and thermal conductivity are included for the transport variables μ and k [3]. The system is closed using the SRK EoS [15] to relate pressure, density, and temperature as follows:

$$p = \frac{RT}{V_m - b} - \frac{a\alpha}{V_m(V_m + b)} \tag{2.3}$$

$$a = \frac{0.42747R^2 T_c^2}{P_c} \tag{2.4}$$

$$b = \frac{0.08664RT_c}{P_c} \tag{2.5}$$

$$\alpha = \left(1 + \left(0.48508 + 1.55171\omega - 0.15613\omega^2\right)\left(1 - T_r^{0.5}\right)\right)^2 \tag{2.6}$$

$$T_r = \frac{T}{T_c} \tag{2.7}$$

where R is the ideal gas constant, T is the absolute temperature, V_m is the molar volume of the species, T_c and P_c are the critical temperature and pressure of the species, respectively, and ω is the acentric factor of the species. Additionally, a, b, and α are all species-specific parameters calculated via equations 2.4, 2.5, and 2.6. All cases are run with a single species, that being CO2.

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BIOGRAPHICAL SKETCH

This is my biography.